THERMAL-NEUTRON MULTIPLE SCATTERING:

CRITICAL DOUBLE SCATTERING

A THESIS

Presented to

The Faculty of the Division of Graduate

Studies and Research

By

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In Partial Fulfillment

of the Requirements for the Degree

Doctor of Philosophy

in the School of Physics

Georgia Institute of Technology

August, 1976

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Date approved by Chairman: 19 Aug 1976

To my best friend, Kathleen

ACKNOWLEDGMENTS

I would like to express my appreciation to my advisor, Dr. Harold A. Gersch, for the help and guidance he provided throughout this research. His vast knowledge of physics, his physical insight and his desire to better understand physical reality make Dr. Gersch an outstanding physicist and advisor.

I would also like to thank Dr. Ronald F. Fox and Dr. Stephen Spooner for serving on the reading committee.

I do not forget my parents, whose initial support and continuing encouragement made my formal education possible.

Finally, and most importantly, for the friendship, encouragement, comfort and love she provided throughout my graduate career, I reserve my deepest appreciation and love for my wife, and best friend, Kathleen.

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SUMMARY

A quantum mechanical formulation for multiple scattering of thermal-neutrons from macroscopic targets is presented and applied to single and double scattering. Critical nuclear scattering from liquids and critical magnetic scattering from ferromagnets are treated in detail in the quasielastic approximation for target systems slightly above their critical points. Numerical estimates are made of the double scattering contribution to the critical magnetic cross section using relevant parameters from actual experiments performed on various ferromagnets. The effect is to alter the usual Lorentzian line shape dependence on neutron wave vector transfer. Comparison with corresponding deviations in line shape resulting from the use of Fisher's modified form of the Ornstein-Zernike spin correlations within the framework of single scattering theory leads to values for the critical exponent η of the modified correlations which reproduce the effect of double scattering.

In addition, it is shown that by restricting the range of applicability of the multiple scattering theory from the outset to critical scattering, Glauber's high energy approximation can be used to provide a much simpler and more powerful description of multiple scattering effects. When sufficiently close to the critical point, it provides a closed form expression for the differential cross section which includes all orders of scattering and has the same form as the single scattering cross section with a modified exponent for the wave vector transfer.

CHAPTER I

INTRODUCTION

Thermal-neutron scattering is one of the most powerful experimental tools for investigating the atomic structure and dynamics of materials. The primary quantity of interest in thermal-neutron scattering from macroscopic materials is the dynamic structure factor, $S(\vec{k},\omega)$, which can be determined from the first Born or single scattering cross section for a neutron to scatter with momentum and energy transfers to the material of $\hbar\vec{k}$ and $\hbar\omega$ respectively. For nuclear scattering of neutrons from liquids, $S(\vec{k},\omega)$ is the Fourier transform of the space and time dependent density-density correlation function, and for magnetic scattering of neutrons from magnetic materials, $S(\vec{k},\omega)$ is the Fourier transform of the space and time dependent spin-spin correlation function. These correlation functions can be determined theoretically from microscopic models of the scattering system from statistical physics.

The experimentally observed scattering cross section is linearly proportional to $S(\vec{k},\omega)$, but only provided that multiple scatterings are negligible. Therefore, the experimental physicist endeavors to make experimental conditions such that multiple scattering is a minimum. However, it is often difficult to experimentally determine if, and to what extent, multiple scattering is present. In addition, while multiple scattering may be small, it is often not negligible and the observed

scattering cross section is not simply proportional to $S(\vec{k},\omega)$. Thus, it is desirable to have a multiple scattering theory to analyze experimental data and from which to quantitatively determine, from experimental parameters, the magnitude of the multiple scattering. Thermal-neutron multiple scattering theories based on classical kinetic theories have been previously obtained.[1,2] In this treatment, a general quantum mechanical formulation for multiple scattering of thermal-neutrons is presented and applied to single and double scattering.

In Chapter II, the general scattering problem is considered and the scattering cross section is introduced. The cross section is expressed in terms of the transition probability for a probe particle to make a transition from a given initial state to a final state. This probability must be modified to describe a scattering process. The remainder of this treatment is specialized to neutron scattering from macroscopic materials.

In Chapter III, the scattering-modified neutron transition probability is determined in a form in which target operators appear only as averages over initial target states. In addition, the modifications made to the probability in order to describe a scattering process are considered in detail.

In Chapter IV, the cross section obtained in Chapter III is expanded in a perturbative series through terms quadrilinear in the interaction. The first term in the series expansion represents single scattering and is shown to yield the time dependent correlation form for the single scattering cross section first given by Van Hove.[3] The

remainder of the terms represent (1) mixed effects of single scattering, refraction and extinction, which are neglected and (2) double scattering. The double scattering term contains the average over initial target states of four target operators, i.e., a four target operator correlation function. For experimental conditions where double scattering is a small effect, the four operator correlation function can, to a good approximation, be decomposed into products of pair correlations. The double scattering can then be expressed in terms of products of two dynamic structure factors. Both the single and double scattering cross sections are expressed in forms applicable to the nuclear and magnetic scattering of neutrons.

In Chapter V, critical nuclear single scattering from liquids is treated in the quasielastic approximation and the classical Ornstein-Zernike (OZ) correlation function,[4] valid for systems near their critical point, is derived using the methods of Klein and Tisza.[5]

In Chapter VI, critical nuclear double scattering from liquids is treated in the quasielastic approximation. The decomposition of the four operator correlation function given in Chapter IV is shown to become virtually exact for small-angle, thermal-neutron critical scattering from targets slightly above their critical point. A double scattering parameter, β, is found from which an estimate of the magnitude of the double scattering can be obtained.

The next two chapters, Chapter VII and VIII, treat, respectively, critical magnetic single and double scattering of unpolarized neutrons from ferromagnets slightly above their Curie temperature. The

scattering is assumed to be quasielastic, and the Heisenberg model of ferromagnetism is adopted for the ferromagnets. The single scattering cross section is determined in a form given by Van Hove.[6] With the appropriate approximations, it is shown that the critical magnetic single and double scattering cross sections can be determined from the critical nuclear single and double scattering cross sections by replacing the nuclear scattering length with the appropriate magnetic scattering length.

In Chapter IX, numerical estimates are made for the double scattering contribution to the critical magnetic cross section using relevant parameters from actual experiments performed on various ferromagnets. Experimental results are often interpreted under the assumption that multiple scattering is negligible, implying that the cross section is proportional to $S(\vec{k},\omega)$. Observed discrepancies from these results have been taken as indicating a failure of the OZ theory. To remove these discrepancies, Fisher proposed a modification to the OZ theory,[7] also discussed in Chapter IX, which in essence involves introducing a new critical exponent, η. This modified theory, when inserted in the single scattering cross section, produces a deviation from the usual Lorentzian line shape of quasielastically scattered neutrons as a function of wave vector transfer. In Chapter IX, it is shown that a similar qualitative effect is produced with double scattering included, but with no modification to the OZ theory, and a functional relationship is obtained between the double scattering parameter, β, and an apparent, double scattering induced η.

In Chapter X, it is shown that by restricting the range of applicability of the multiple scattering theory presented in Chapters III and IV from the outset to critical scattering, Glauber's high energy approximation[8] can be used to provide a much simpler and more powerful description of multiple scattering effects. The Glauber approximation is shown to give exactly the same results for critical scattering as obtained in Chapters V through VIII from first principles. In addition, when sufficiently close to the critical point, a closed form expression is obtained for the cross section which includes all orders of scattering and has the same form as the single scattering cross section with a modified exponent for the wave vector transfer.

The limits of integration on the various integrals appearing in this work have been explicitly displayed only when their omission might prove to be a source of confusion. Also, the Einstein convention of summing over repeated indices has been assumed.

CHAPTER II

THE SCATTERING PROBLEM

Scattering enjoys a very prominent position in both theoretical and experimental physics. It serves as the most powerful tool used by the experimental physicist in the investigation of microscopic phenomena in matter. Inaccessible to direct measurement, the properties of the microscopic world must be deduced from indirect measurements, such as those in a scattering experiment. In such experiments, probe particles prepared in a known initial state are scattered from a target of interest also prepared in a known initial state or a known distribution of states. The interaction between the probe particle and the target causes transitions in both the probe and the target. If this interaction is precisely known and is sufficiently weak, knowledge can be gained about the target from measurements of the final probe particle states.

The type of probe used and the initial state in which it is prepared will depend on the target and on the physical quantities to be measured in the target. For example, if one wanted to measure the charge distribution in nuclei, one would need a probe particle that possesses a charge, so that it would interact with the charge distribution in the nucleus. In addition, the probe would have to have a de Broglie wavelength comparable to the dimension of the nucleus, so

that quantum interference effects would be present. High energy electrons would be an appropriate probe.

Thermal-neutrons are especially well suited as probes in determining the structure and dynamics of condensed matter, i.e., liquids and solids. The neutron, being a neutral particle with a magnetic moment, will be basically involved in two types of interactions with the target: nuclear and magnetic. Since thermal-neutrons have wavelengths comparable to the separation of the atoms in condensed matter, they can be used to determine physical quantities like density-density correlations, via nuclear interactions, and spin-spin correlations, via magnetic interactions. In addition, several simplifing features occur with the use of thermal-neutrons. For example, due to the thermal-neutron's low energy content, its wavelength is much longer than the range of the nuclear interaction between the neutron and a target nucleus. Therefore, the true nuclear interaction can be replaced with its S-wave approximation, i.e., the Fermi-pseudo potential. When considering magnetic interactions with the electrons of the target, electronic excitations can be neglected, again, due to the thermal-neutron's low energy content. Also, since the neutron is a neutral particle, no complicated long-ranged Coulombic interactions are present. Put simply, thermal-neutrons as probes are simple enough to be handled with some mathematical rigor, yet, rich enough to still be physically interesting.

The physical quantity of primary interest measured by an experimentalist is the scattering cross section. Therefore, the theoretician should express his results in terms of the cross section. The cross

section, $d\sigma$, is defined as the number of probe particles scattered into a solid angle $d\Omega$ per unit time, $\frac{dN_{sc}}{dt}$, divided by the number of incident probe particles per unit area per unit time, $\frac{1}{A}\frac{dN_{inc}}{dt}$,

$$d\sigma = \frac{dN_{sc}/dt}{A^{-1}\, dN_{inc}/dt} \tag{2.1}$$

The cross section is independent of the intensity of the incoming beam of probe particles; therefore, letting the beam consist of one probe particle will not alter the resulting cross section. The denominator of the cross section given by Eq. (2.1) may be written as

$$\frac{1}{A}\frac{dN_{inc}}{dt} = j_{inc} \tag{2.2}$$

where j_{inc} is the probability current density for the incoming probe particle.

The probe, before and after interacting with the target, is a free particle whose stationary state wavefunction, $\langle \vec{r} | \vec{k} \rangle = \psi_{\vec{k}}(\vec{r})$, is determined from the time independent Schrodinger equation,

$$H_p \psi_{\vec{k}}(\vec{r}) = E_k \psi_{\vec{k}}(\vec{r}) \tag{2.3}$$

where H_p is the free particle Hamiltonian,

$$H_p = -\frac{\hbar^2\nabla^2}{2m} \tag{2.4}$$

and m is the mass of the probe. Writing

$$E_k = \frac{\hbar^2 k^2}{2m} \tag{2.5}$$

and using Eq. (2.4), Schrodinger's equation becomes

$$(\nabla^2 + k^2)\psi_{\vec{k}}(\vec{r}) = 0. \tag{2.6}$$

A plane wave solution of Eq. (2.6) is

$$\psi_{\vec{k}}(\vec{r}) = Ce^{-i\vec{k}\cdot\vec{r}} \tag{2.7}$$

where the normalization constant, C, is to be determined by the imposed boundary conditions.

Using the wavefunction given by Eq. (2.7) to describe the probe would lead to the non-physical result that the probability density, ρ, of finding the probe at any point $\vec{r}$ is constant. This fact results from the definition of ρ, i.e.,

$$\rho \equiv \left|\psi_{\vec{k}}(\vec{r})\right|^2 = \left|C\right|^2 \tag{2.8}$$

Therefore, the probe has an equal probability of being found at all points in space. To localize the probe in space in order to reflect laboratory conditions, a wave packet should be formed by taking an appropriate superposition of wavefunctions given by Eq. (2.7), i.e.,

$$\psi(\vec{r}) = \sum_{\vec{k}} c_{\vec{k}} \psi_{\vec{k}}(\vec{r}) \tag{2.9}$$

If the wavefunction given by Eq. (2.9) is taken to describe the wave packet at the initial time t = 0, then the time evolvement of the wave packet, while it represents the probe as a free particle, is governed by the time dependent Schrodinger equation,

$$H_p \psi(\vec{r},t) = i\hbar \frac{\partial \psi(\vec{r},t)}{\partial t} \tag{2.10}$$

The solution to Eq. (2.10) is

$$\psi(\vec{r},t) = \sum_{\vec{k}} c_{\vec{k}} \psi_{\vec{k}}(\vec{r}) \; e^{-iE_k t/\hbar} \tag{2.11}$$

This, indeed, is one way to proceed in solving the scattering problem.[9] However, it is possible to use the simple stationary state wavefunction, Eq. (2.7), instead of the more complicated time dependent wave packet, Eq. (2.11), to describe the probe. To do so, appropriate adjustments must be made later on in the theory to reflect the fact that the probe approaches, interacts, then leaves the target. These adjustments are

discussed in detail in Chapter III.

In order to construct a complete set of orthonormal probe wavefunctions of the type given by Eq. (2.7), i.e., wavefunctions that satisfy

$$\int d^3r \; \psi_{\vec{k}}(\vec{r})\psi_{\vec{k}'}(\vec{r}) = \delta_{\vec{k},\vec{k}'} \tag{2.12}$$

a purely mathematical set of boundary conditions are imposed on the wavefunctions. The probe and target are enclosed in an imaginary box of arbitrarily large volume $V = L^3$ and periodic boundary conditions are imposed. That is, an infinite number of boxes, side by side, containing the probe and target are envisioned, and the wavefunction is required to have the same value at corresponding space points in the various boxes, i.e.,

$$\psi_{\vec{k}}(x,y,z) = \psi_{\vec{k}}(x + n_x L,\; y + n_y L,\; z + n_z L) \tag{2.13}$$

where n_x, n_y and n_z can take on all integer values. In order to satisfy Eq. (2.13), the components of $\vec{k}$ must be restricted to take on certain discrete values. In rectangular coordinates, these values are

$$k_i = \frac{n_i 2\pi}{L} \tag{2.14}$$

where $i = x,y,z$. Now, requiring the wavefunctions to satisfy Eq. (2.12) when the integration goes over the volume of any one of the boxes

gives for the normalization constant in Eq. (2.7),

$$C = V^{-\frac{1}{2}} \tag{2.15}$$

Thus from Eqs. (2.8) and (2.15), the probability density of the probe is given by

$$\rho = V^{-1} \tag{2.16}$$

Assuming that the probe is initially prepared in an eigenstate $|\vec{k}_i\rangle$ of H_p with velocity $\vec{v}_i$, the probability current density is

$$j_{inc} = \rho v_i = \frac{\hbar k_i}{mV} \tag{2.17}$$

The numerator, $\frac{dN_{sc}}{dt}$, of the cross section given by Eq. (2.1) is related to the time rate of change of the modified probability, P_s, that the scattering system (probe and target) will make a transition from the given prepared initial state at time t_o to a final state $|T_f\rangle|\vec{k}_f\rangle$ at some later time t. Here $|T_f\rangle$ denotes a state of the target. The probability must be modified to describe a scattering process (see Chapter III). Since only the final states of the probe are measured, regardless of the final target states, the final target states must be summed over. In some scattering processes, e.g., atomic collisions, it is sometimes possible to explicitly insert the final target states in the summing process, due to the manageable (small) number of such

states. However, when the target is a many-body, macroscopic system such as a solid or liquid, the huge number of possible final target states prohibits this procedure. Thus, the hope is that the sum over these final target states as they appear in the probability, P_S, can be removed due to closure.

The final state of the probe, $|\vec{k}_f\rangle$, is specified by the vector quantum "number" $\vec{k}_f$. Since $\vec{k}_f$ is a vector, $|\vec{k}_f\rangle$ is actually specified by three quantum numbers. In rectangular coordinates, these three numbers are k_x, k_y and k_z. In spherical polar coordinates, they are k_f, θ and ϕ. Since the probe is a free particle, its energy, E_f, is given by

$$E_f = \frac{\hbar^2 k_f^2}{2m} \tag{2.18}$$

Therefore, an equally good set of quantum numbers to specify $|\vec{k}_f\rangle$ are E_f, θ and ϕ. Thus, a final probe state is specified by its energy and the orientation in space of the wave number vector, $\vec{k}_f$. Now, since $\frac{dN_{sc}}{dt}$ is the number of probe particles scattered into a solid angle $d\Omega = \sin\theta\, d\theta\, d\phi$ per unit time regardless of the energies of the probe particles, and since only one incident probe particle is being considered,

$$\frac{dN_{sc}}{dt} = \sum_{T_f} \int_{E_f} \frac{dP_S}{dt} \rho(E_f) dE_f d\Omega \tag{2.19}$$

where $\rho(E_f)dE_f d\Omega$ is the number of final probe states in a solid angle

$d\Omega$ with energies between E_f and $E_f + dE_f$, where the final probe energies have been integrated over and where the final states of the target appearing in P_S have been summed over. If k_f, θ and ϕ are chosen as the quantum numbers of $|\vec{k}_f\rangle$, then the density of final states is given by $\rho(\vec{k}_f)d^3k_f$ and

$$\rho(E_f)dE_f d\Omega = \rho(\vec{k}_f)d^3k_f = \rho(\vec{k}_f)k_f^2 dk_f d\Omega \tag{2.20}$$

The density of final probe states, $\rho(\vec{k}_f)$, can be determined by considering $\vec{k}$-space. From Eq. (2.14), every final state of the probe is represented by a point in $\vec{k}$-space with coordinates $\left(\frac{n_x 2\pi}{L}, \frac{n_y 2\pi}{L}, \frac{n_z 2\pi}{L}\right)$. Therefore, each point occupies a volume of $\left(\frac{2\pi}{L}\right)^3$ in $\vec{k}$-space, giving for the density

$$\rho(\vec{k}_f) = \frac{V}{(2\pi)^3} \tag{2.21}$$

Now, from Eq. (2.20)

$$\rho(E_f) = \rho(\vec{k}_f)k_f^2 \frac{dk_f}{dE_f} \tag{2.22}$$

and from Eq. (2.18)

$$\frac{dk_f}{dE_f} = \frac{m}{\hbar^2 k_f} \tag{2.23}$$

Therefore, the density of final states is

$$\rho(E_f) = \frac{V}{(2\pi)^3}\frac{k_f m}{\hbar^2} \tag{2.24}$$

From Eqs. (2.2), (2.17), (2.19) and (2.24), the cross section, Eq. (2.1), is now given by

$$d\sigma = \frac{V^2 m^2}{(2\pi\hbar)^3}\sum_{T_f}\int_{E_f}\frac{k_f}{k_i}\frac{dP_S}{dt}\,dE_f d\Omega \tag{2.25}$$

The quantity often measured in the laboratory is the differential cross section (DCS), $\frac{d\sigma}{d\Omega}$, which is readily obtained from Eq. (2.25),

$$\frac{d\sigma}{d\Omega} = \frac{V^2 m^2}{(2\pi\hbar)^3}\sum_{T_f}\int_{E_f}\frac{k_f}{k_i}\frac{dP_S}{dt}\,dE_f \tag{2.26}$$

With the sophisticated equipment available to the experimental physicist today, the scattered probe particles can be energy analyzed; thus the partial or double differential cross section (DDCS), $\frac{d^2\sigma}{d\Omega dE_f}$, can be measured. From Eq. (2.26), the DDCS is given by

$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{V^2 m^2}{(2\pi\hbar)^3}\frac{k_f}{k_i}\sum_{T_f}\frac{dP_S}{dt} \tag{2.27}$$

The appearance of the quantization volume, V, in the cross section, Eqs. (2.26) and (2.27), is due to the artificial boundary conditions imposed to produce normalized probe states. Certainly, no measurable physical quantity can depend on such an arbitrary, non-physical quantity. As it turns out, the probability P_S, goes like V^{-2}, so that the V's will cancel. Since all measurable physical quantities

must and will be independent of V, this quantization volume will be set equal to unity from now on wherever it appears.

The theoretical scattering problem to be attacked and solved is now clear. It is, for a given target and probe, to determine the scattering-modified probe transition probability, $\sum_{T_f} P_S$, in a form useful for the analysis of experimental data. The next chapter will be devoted to this task. In the remainder of this treatment, only the case where the probe is a thermal-neutron and the target is a macroscopic sample of condensed matter, i.e., a liquid or a solid, will be considered.

CHAPTER III

THE SCATTERING-MODIFIED NEUTRON TRANSITION PROBABILITY

The DDCS for thermal-neutron scattering was determined in Chapter II and is given by Eq. (2.27)

$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{m_N^2}{(2\pi\hbar)^3}\frac{k_f}{k_i}\frac{dP_N}{dt} \qquad (V=1) \tag{3.1}$$

where m_N is the neutron mass and

$$P_N = \sum_{T_f} P_S \tag{3.2}$$

is the neutron transition probability, modified to describe a scattering process. In this chapter, the scattering-modified neutron transition probability is determined in a form in which the target operators appear only as averages over initial target states.

The Hamiltonian, H, for the scattering system is given by

$$H = H_N + H_T + \phi(A,B) \tag{3.3}$$

where H_N is the Hamiltonian of the neutron, H_T is the Hamiltonian of the target, ϕ represents the interaction between the neutron and the target and A and B represent some dynamical operators of the neutron and target respectively. It is assumed that the neutron is initially

prepared in a plane wave eigenstate, $|\vec{k}_i\rangle$, of H_N. A practical difficulty arises in the initial preparation of the target. Since the target is taken to be a macroscopic sample, it is generally impossible to prepare it in a pure state; thus it cannot be represented by a state vector. The best that can be done, is to prepare the target in a known distribution of states, i.e., a mixed state. Therefore, it is natural, although not necessary, to describe the scattering system in density operator language, which treats both pure states and mixed states on an equal basis.

Assuming that the neutron and the target begin interacting with each other at time t_o, the scattering system density operator, $\rho_S(t)$, can be written, at time t_o, as a product of the neutron density operator, $\rho_N(t)$, at time t_o and the target density operator, $\rho_T(t)$, at time t_o,

$$\rho_S(t_o) = \rho_N(t_o)\rho_T(t_o) \tag{3.4}$$

where

$$\rho_N(t_o) = |\vec{k}_i\rangle\langle\vec{k}_i| \tag{3.5}$$

In terms of the eigenstates, $|T_i\rangle$, of H_T, the initial target density operator can be written

$$\rho_T(t_o) = \sum_{T_i} P_{T_i} |T_i\rangle\langle T_i| \tag{3.6}$$

where P_{T_i} is the probability of the occurrence of the target state $|T_i\rangle$. From Eqs. (3.5) and (3.6), the initial scattering system density operator, Eq. (3.4), is

$$\rho_S(t_o) = \sum_{T_i} P_{T_i} |T_i\rangle|\vec{k}_i\rangle\langle\vec{k}_i|\langle T_i| \tag{3.7}$$

The time evolvement of $\rho_S(t)$ is determined from the equation of motion,

$$i\hbar \frac{d\rho_S(t)}{dt} = [H,\rho_S(t)] \tag{3.8}$$

where $[H,\rho] = H\rho - \rho H$. Formally solving Eq. (3.8) gives

$$\rho_S(t) = e^{-iH(t - t_o)/\hbar} \rho_S(t_o)\, e^{iH(t - t_o)/\hbar} \tag{3.9}$$

Therefore, from Eq. (3.7), the scattering system density operator at time t, Eq. (3.9), is

$$\rho_S(t) = \sum_{T_i} P_{T_i}\, e^{-iH(t - t_o)/\hbar} |T_i\rangle|\vec{k}_i\rangle\langle\vec{k}_i|\langle T_i|\, e^{iH(t - t_o)/\hbar} \tag{3.10}$$

The probability, $P'_S(t)$, that the scattering system will be found in eigenstate, $|\vec{k}_f\rangle|T_f\rangle$, of $H_N + H_T$ at time t is given by

$$P'_S(t) = \langle T_f|\langle\vec{k}_f|\rho_S(t)|\vec{k}_f\rangle|T_f\rangle \tag{3.11}$$

where the prime on $P'_S(t)$ indicates that the probability has not yet been modified to describe a scattering process. From Eqs. (3.10) and (3.11), the neutron transition probability is

$$P'_N(t) = \sum_{T_f} P'_S(t) = \sum_{T_f} \sum_{T_i} P_{T_i} \langle T_f | \langle \vec{k}_f | \, e^{-iH(t - t_o)/\hbar} \, | T_i \rangle | \vec{k}_i \rangle$$

$$\times \langle \vec{k}_i | \langle T_i | \, e^{iH(t - t_o)/\hbar} \, | \vec{k}_f \rangle | T_f \rangle \tag{3.12}$$

The exponentials in Eq. (3.12) are, from Eq. (3.3),

$$e^{\pm iH(t - t_o)/\hbar} = e^{\pm i[H_N + H_T + \phi](t - t_o)/\hbar} \tag{3.13}$$

Since ϕ does not commute with either H_N or H_T, these exponentials cannot be written as a simple product of exponentials. However, using the appropriate time ordering operators, one can write

$$e^{-iH(t - t_o)/\hbar} = e^{-i(H_N + H_T)t/\hbar} \left[T_+ e^{-\frac{i}{\hbar} \int_{t_o}^{t} dt' \phi(t')} \right]$$

$$\times e^{i(H_N + H_T)t_o/\hbar} \tag{3.14}$$

$$e^{iH(t - t_o)/\hbar} = e^{-i(H_N + H_T)t_o/\hbar} \left[T_- e^{\frac{i}{\hbar}\int_{t_o}^{t} dt'\phi(t')} \right]$$

$$\times e^{i(H_N + H_T)t/\hbar} \tag{3.15}$$

where $\phi(t)$ is in the interaction picture,

$$\phi(t) = e^{i(H_N + H_T)t/\hbar}\, \phi\, e^{-i(H_N + H_T)t/\hbar} \tag{3.16}$$

and the operator $T_+(T_-)$ time orders products of the $\phi(t)$'s with the earlier (later) time to the right. A derivation of the identities given by Eqs. (3.14) and (3.15) is provided in Appendix I. Inserting these identities into Eq. (3.12), rearranging the matrix elements and using closure over the final target states, i.e.,

$$\sum_{T_f} |T_f\rangle\langle T_f| = 1 \tag{3.17}$$

the neutron transition probability becomes,

$$P'_N(t) = \sum_{T_i} P_{T_i} \langle \vec{k}_i | \langle T_i | T_- e^{\frac{i}{\hbar}\int_{t_o}^{t} dt'\phi(t')} | \vec{k}_f \rangle$$

$$\times \langle \vec{k}_f | T_+ e^{-\frac{i}{\hbar}\int_{t_o}^{t} dt'\phi(t')} | T_i \rangle | \vec{k}_i \rangle \tag{3.18}$$

At this point, the neutron transition probability, $P_N'(t)$, given by Eq. (3.18) is modified to describe a scattering process. The fact that $P_N'(t)$ does not describe a scattering process can readily be seen by noting that, in general, $\frac{dP_N'(t)}{dt}$ is time dependent; thus, from Eq. (3.1), it would give rise to a nonsensical time dependent DDCS. The neutron transition probability does describe the unphysical situation of suddenly immersing the target, at time t_o, in a plane wave, which represents the neutron. Now, since a plane wave has a non-zero amplitude at all points in space, the target and neutron will interact indefinitely from time t_o. In such a process, $\frac{dP_N'(t)}{dt}$ will surely depend on time.

In a scattering process, the neutron and target will not interact when they are sufficiently far apart at some initial time t_o. As time evolves from t_o and the neutron moves closer to the target, the strength of the interaction, ϕ, will continuously increase from zero to its full value when the neutron is on top of the target, then continuously decrease to zero again as the neutron leaves the target. This scattering process would be described by the formalism if the neutron were represented by a wave packet since, then, the neutron would be localized in space and the strength of the interaction, ϕ, would be governed by the functional form of ϕ as it depends on the relative neutron-target separation.

The description of the scattering process is lost by using non-localized plane waves to represent the neutron, but can be regained if

the interaction is artificially turned on and turned off in time to mimic the passing of the target by the neutron in space. Without loss of generality, the time t = 0 is chosen for the time when the neutron and target are fully interacting, and the period of effective interaction is denoted by $\Delta\tau$. Now, taking the initial time t_o to be in the remote past, i.e. $t_o \to -\infty$, the interaction, ϕ, is modified thusly,

$$\phi(t) \to \phi(t)\ e^{t/\Delta\tau} \qquad (t \lesssim 0) \tag{3.19}$$

The corresponding modification of $\phi(t)$ for times $t \gtrsim 0$ need not be considered; for, clearly, the neutron transition rate, $\frac{dP_N}{dt}$, as it appears in the DDCS Eq. (3.1), describes a scattering process only for times

$$|t| << \Delta\tau \tag{3.20}$$

since $\frac{dP_N}{dt} \cong 0$ for times $|t| \gtrsim \Delta\tau$. Once the modification (3.19) of $\phi(t)$ has been made, the limit $t_o \to -\infty$ imposed and all arithmetic manipulations have been completed, $\Delta\tau$ can be removed from the resulting expressions, having served its purpose, by letting $\frac{1}{\Delta\tau} \to 0$. Physically, letting $\frac{1}{\Delta\tau} \to 0$ is a statement of the fact that the uncertainty in the interaction time is large. One consequence of this, as will be shown, is that the uncertainty in the scattering system's energy is small, i.e., energy is conserved. This is, of course, in agreement with the uncertainty principle, i.e.,

$$\Delta E \Delta t \gtrsim \hbar \tag{3.21}$$

One additional modification is necessary. Since the cross section is defined in terms of the number of neutrons scattered, i.e., the number of neutrons that actually interact with the target, $P'_N(t)$ must be modified to exclude the probability of a neutron passing straight through the target without ever having interacted with the target. That is, the neutron transition probability must be modified so that it vanishes as $\phi(t)$ vanishes. This is accomplished by simply subtracting the number one from both exponentials in $P'_N(t)$, Eq. (3.18).

The above mentioned modifications are now applied to the neutron transition probability, $P'_N(t)$, Eq. (3.18), in order to obtain the scattering-modified neutron transition probability, $P_N(t)$. Thus,

$$P_N(t) = \lim_{\varepsilon \to 0} \sum_{T_i} P_{T_i} \langle \vec{k}_i | \langle T_i | T_- \exp\left[\frac{i}{\hbar} \int_{-\infty}^{t} dt' \phi(t') e^{\varepsilon t'} \right] - 1 | \vec{k}_f \rangle$$

$$\times \langle \vec{k}_f | T_+ \exp\left[- \frac{i}{\hbar} \int_{-\infty}^{t} dt' \phi(t') e^{\varepsilon t'} \right] - 1 | T_i \rangle | \vec{k}_i \rangle \tag{3.22}$$

where

$$\varepsilon \equiv \frac{1}{\Delta \tau} \tag{3.23}$$

As will be shown, the time derivative of $P_N(t)$ given by Eq. (3.22)

will indeed be independent of time when the time restriction (3.20) is properly taken into account. From Eq. (3.23), this restriction is now

$$\varepsilon|t| \ll 1 \tag{3.24}$$

In order to obtain $P_N(t)$ in a form in which target operators appear only as averages over initial target states, the final neutron state vectors in Eq. (3.22) must be removed from their present position "inside" the target average. Formally this can be accomplished, even though $\phi(t)$ contains neutron operators A, by introducing the subscripts 1,2 on neutron operators and state vectors, by writing Eq. (3.22) as

$$P_N(t) = \lim_{\varepsilon\to 0} \sum_{T_i} P_{T_i} \langle \vec{k}_{i_1} | \langle T_i | T_- \exp\left[\frac{i}{\hbar}\int_{-\infty}^{t} dt' \phi_1(t') e^{\varepsilon t'}\right] - 1 | \vec{k}_{f_1} \rangle$$

$$\times \langle \vec{k}_{f_2} | T_+ \exp\left[-\frac{i}{\hbar}\int_{-\infty}^{t} dt' \phi_2(t') e^{\varepsilon t'}\right] - 1 | T_i \rangle | \vec{k}_{i_2} \rangle \tag{3.25}$$

and by introducing a new position ordering operator, P_{12}, which moves all operators and state vectors with subscript 1 to the left of all operators and state vectors with subscript 2. Introducing this operator into Eq. (3.25) allows one to freely move all operators and state vectors with different subscripts past each other, since P_{12} will restore the correct order. Therefore,

$$P_N(t) = \lim_{\varepsilon \to 0} P_{12} \langle \vec{k}_{f_2} | \langle \vec{k}_{i_1} | \langle (T_- \exp \left[\frac{i}{\hbar} \int_{-\infty}^{t} dt' \phi_1(t') e^{\varepsilon t'} \right] - 1)$$

$$\times (T_+ \exp \left[- \frac{i}{\hbar} \int_{-\infty}^{t} dt' \phi_2(t') e^{\varepsilon t'} \right] - 1) \rangle | \vec{k}_{i_2} \rangle | \vec{k}_{f_1} \rangle \qquad (3.26)$$

where

$$\langle \ldots \rangle = \sum_{T_i} P_{T_i} \langle T_i | \ldots | T_i \rangle \, , \qquad (3.27)$$

$$\phi_1(t) = \phi[A_1(t), B(t)] \qquad (3.28)$$

and

$$A_1(t) = e^{iH_{N_1} t/\hbar} \, A_1 \, e^{-iH_{N_1} t/\hbar} \qquad (3.29)$$

Once the positioning operator P_{12} has restored the correct ordering among the neutron operators and state vectors, the subscripts 1,2 may be dropped.

Therefore, at least formally, the scattering-modified neutron transition probability, $P_N(t)$, as given by Eq. (3.26), is now in a form in which target operators appear only as averages over initial target states. Differentiating $P_N(t)$ with respect to time, then inserting the results into Eq. (3.1) gives the DDCS in closed form. However, this resulting expression for the DDCS is of little practical utility

due to the presence of the time and position ordering operators. Obtaining the cross section in a closed form useful for analysis of experimental data appears, at best, to be a formidable task, and will be considered briefly in Chapter X within the framework of critical scattering and then only in an approximate way. A perturbative expansion of the DDCS through second order will be considered in the next chapter, and in this regard, the formalism presented here appears to be more powerful than other known formalisms when dealing with macroscopic targets. For example, the cross section in terms of the transition matrix can also be written in closed form, again, with little practical utility. Upon expanding in a perturbative series, the first order term explicitly contains final target states, while higher order terms contain both final target states and sums over intermediate target states. By adroit mathematical maneuvering, Van Hove[3] showed how, in first order, the sum over final target states could be removed from the cross section via closure, thus leaving the cross section in terms of the dynamic structure factor, $S(\vec{k},\omega)$, which contains target operators only as averages over initial target states, i.e., time-dependent correlation functions of target operators. Removing the intermediate and final target states in higher order terms in order to obtain the cross section in terms of correlation functions appears to require even more mathematical ingenuity. However, in the formalism presented here, the DDCS, in closed form, is already expressed in terms of the initial target average of target operators; thus all terms in a perturbative expansion will be expressed in terms of correlation functions of target operators.

An expansion of the cross section in which every term is expressed in terms of correlation functions of target operators is and must be equivalent to any other correctly done expansion. The practical utility of the former expansion is that correlation functions have a well known physical meaning and can be obtained exactly or approximately from microscopic models in statistical physics. Herein lies the power of the formalism presented in this chapter.

CHAPTER IV

THERMAL-NEUTRON SINGLE AND DOUBLE SCATTERING

In this chapter, the DDCS obtained in Chapter III is expanded in a perturbative series and the single and double scattering cross sections are considered in detail.

The DDCS for thermal-neutron scattering was obtained in Chapter III and is given by

$$\frac{d^2\sigma}{d\Omega dE_f} = \lim_{\varepsilon\to 0} \frac{m_N^2}{(2\pi\hbar)^3} \frac{k_f}{k_i}$$

$$\times P_{12} \frac{d}{dt} \langle \vec{k}_{f_2} | \langle \vec{k}_{i_1} | \langle (T_- \exp\left[\frac{1}{\hbar} \int_{-\infty}^{t} dt' \phi_1(t') \, e^{\varepsilon t'} \right] - 1)$$

$$\times (T_+ \exp\left[-\frac{1}{\hbar} \int_{-\infty}^{t} dt' \phi_2(t') \, e^{\varepsilon t'} \right] - 1) \rangle | \vec{k}_{i_2} \rangle | \vec{k}_{f_1} \rangle \quad (4.1)$$

where the target operators, A(t), which are contained in the interaction $\phi(t)$, appear only in the average over initial target states. To proceed further, a specific form for the interaction is chosen, namely,

$$\phi(t) = A(t)B(t). \quad (4.2)$$

This form for the interaction is found in many processes, including the

nuclear and magnetic scattering of neutrons (see Chapters V and VII). Now, since A_1, A_2 and B all commute with each other, the time ordering operators appearing in Eq. (4.1) may be written as

$$T_+ = T_+^{A_2}\, T_+^{B}$$
$$T_- = T_-^{A_1}\, T_-^{B} \tag{4.3}$$

Therefore, from Eqs. (4.2) and (4.3), the target average appearing in Eq. (4.1) becomes

$$\langle \ldots \rangle = T_-^{A_1}\, T_+^{A_2} \Big\langle \Big(T_-^{B} \exp\Big[\frac{i}{\hbar}\int_{-\infty}^{t} dt' A_1(t')B(t')\, e^{\varepsilon t'}\Big] - 1\Big)$$
$$\times \Big(T_+^{B} \exp\Big[-\frac{i}{\hbar}\int_{-\infty}^{t} dt' A_2(t')B(t')\, e^{\varepsilon t'}\Big] - 1\Big)\Big\rangle \tag{4.4}$$

In order to obtain the single and double scattering cross sections, Eq. (4.1) must be expanded in a perturbative series. This is accomplished by expanding the target average, Eq. (4.4), in a Taylor's series, i.e.,

$$\Delta(\lambda) \equiv \Big\langle \Big(T_-^{B} \exp\Big[\lambda \frac{i}{\hbar}\int_{-\infty}^{t} dt' A_1(t')B(t')\, e^{\varepsilon t'}\Big] - 1\Big)$$
$$\times \Big(T_+^{B} \exp\Big[-\lambda \frac{i}{\hbar}\int_{-\infty}^{t} dt' A_2(t')B(t')\, e^{\varepsilon t'}\Big] - 1\Big)\Big\rangle = \sum_{n=0}^{\infty} \frac{\Delta_n}{n!}\lambda^n \tag{4.5}$$

where

$$\Delta_n = \left.\frac{d^n \Delta(\lambda)}{d\lambda^n}\right|_{\lambda=0} \tag{4.6}$$

and where finally, λ is set equal to unity. This is equivalent to expanding the exponentials in Eq. (4.4) and writing the results as a power series in B. The first several terms are

$$\Delta_0 = \Delta_1 = 0 \tag{4.7}$$

$$\frac{\Delta_2}{2!} = \frac{1}{\hbar^2} \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t} dt_2 A_1(t_1) A_2(t_2) \langle B(t_1)B(t_2) \rangle \; e^{\varepsilon(t_1 + t_2)} \tag{4.8}$$

$$\begin{aligned}\frac{\Delta_3}{3!} = \frac{i}{\hbar^3} &\int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t} dt_3 A_1(t_1)A_1(t_2)A_2(t_3) \\ &\times \langle B(t_2)B(t_1)B(t_3) \rangle \; e^{\varepsilon(t_1 + t_2 + t_3)} \\ - \frac{i}{\hbar^3} &\int_{-\infty}^{t} dt_1 \int_{-\infty}^{t} dt_2 \int_{-\infty}^{t_2} dt_3 A_1(t_1)A_2(t_2)A_2(t_3) \\ &\times \langle B(t_1)B(t_2)B(t_3) \rangle \; e^{\varepsilon(t_1 + t_2 + t_3)}\end{aligned} \tag{4.9}$$

$$\frac{\Delta_4}{4!} = -\frac{1}{\hbar^4}\int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_2} dt_3 \int_{-\infty}^{t} dt_4 A_1(t_1)A_1(t_2)A_1(t_3)A_2(t_4)$$

$$\times \langle B(t_3)B(t_2)B(t_1)B(t_4)\rangle\, e^{\varepsilon(t_1 + t_2 + t_3 + t_4)}$$

$$+\frac{1}{\hbar^4}\int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t} dt_3 \int_{-\infty}^{t_3} dt_4 A_1(t_1)A_1(t_2)A_2(t_3)A_2(t_4)$$

$$\times \langle B(t_2)B(t_1)B(t_3)B(t_4)\rangle\, e^{\varepsilon(t_1 + t_2 + t_3 + t_4)}$$

$$-\frac{1}{\hbar^4}\int_{-\infty}^{t} dt_1 \int_{-\infty}^{t} dt_2 \int_{-\infty}^{t_2} dt_3 \int_{-\infty}^{t_3} dt_4 A_1(t_1)A_2(t_2)A_2(t_3)A_2(t_4)$$

$$\times \langle B(t_1)B(t_2)B(t_3)B(t_4)\rangle\, e^{\varepsilon(t_1 + t_2 + t_3 + t_4)} \qquad (4.10)$$

The DDCS can now be written as

$$\frac{d^2\sigma}{d\Omega dE_f} = \lim_{\varepsilon\to 0} \frac{m_N^2}{(2\pi\hbar)^3}\frac{k_f}{k_i} P_{12}\frac{d}{dt}\langle \vec{k}_{f_2} | \langle \vec{k}_{i_1} | T_-^{A_1} T_+^{A_2} \sum_{n=2}^{\infty} \frac{\Delta_n}{n!} | \vec{k}_{i_2}\rangle | \vec{k}_{f_1}\rangle$$

(4.11)

The Δ_2 term, Eq. (4.8), represents single scattering and retaining only this term in Eq. (4.11) would result in the first Born scattering cross section. The Δ_3 term, Eq. (4.9), contains the mixed

effects of single scattering and renormalized probe behavior due to the fact that the neutron is moving in the medium of the target, i.e., a refraction effect. The first and last terms in Eq. (4.10) contain the mixed effects of single scattering, refraction, and depletion of the neutron beam due to the fact that scattering is occurring, i.e., an extinction effect. The middle term in Δ_4 contains the true double scattering. This term and Δ_2 will be the only terms retained in order to obtain the double and single scattering cross sections. The neglected terms and the justification for the physical interpretation given them above will be discussed in Chapter X.

Single Scattering

Retaining only the Δ_2 term in the DDCS, Eq. (4.11), results in the first Born or single scattering cross section,

$$\frac{d^2\sigma(1)}{d\Omega dE_f} = \lim_{\varepsilon\to 0} \frac{m_N^2}{(2\pi\hbar)^3} \frac{1}{\hbar^2} \frac{k_f}{k_i}$$

$$\times P_{12} \frac{d}{dt} \langle \vec{k}_{f_2} | \langle \vec{k}_{i_1} | \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t} dt_2 A_1(t_1) A_2(t_2)$$

$$\times \langle B(t_1)B(t_2) \rangle \, e^{\varepsilon(t_1 + t_2)} \, |\vec{k}_{i_2}\rangle |\vec{k}_{f_1}\rangle \qquad (4.12)$$

Equation (4.12) can be transcribed into Van Hove's form for the first Born scattering cross section containing the dynamic structure factor.[3] For an interaction depending on distance only,

$$\phi(t) = \sum_j \phi[\vec{r}(t)] = \sum_{\vec{k}_1} g(\vec{k}_1)\, e^{i\vec{k}_1 \cdot \vec{r}_N(t)} \sum_j e^{-i\vec{k}_1 \cdot \vec{r}_j(t)} \tag{4.13}$$

where $\vec{r}(t) = \vec{r}_N(t) - \vec{r}_j(t)$, $\vec{r}_N(t)$ being the neutron position vector, $\vec{r}_j(t)$ being the position vector of the j^{th} target atom and $g(\vec{k}_1)$ is the Fourier transform of $\phi(\vec{r})$, i.e.,

$$g(\vec{k}_1) = \int d^3r\, e^{-i\vec{k}_1 \cdot \vec{r}}\, \phi(\vec{r}) \tag{4.14}$$

Therefore, the interaction, ϕ, may be written as

$$\phi(t) = \sum_{\vec{k}_1} g(\vec{k}_1) A(\vec{k}_1, t) B(\vec{k}_1, t) \tag{4.15}$$

where

$$A(\vec{k}_1, t) = e^{i\vec{k}_1 \cdot \vec{r}_N(t)} \tag{4.16}$$

$$B(\vec{k}_1, t) = \sum_j e^{-i\vec{k}_1 \cdot \vec{r}_j(t)} \equiv n_{\vec{k}_1}(t) \tag{4.17}$$

and $n_{\vec{k}_1}(t)$ is the Fourier transform of the density operator. The interaction is now in the desired form given by Eq. (4.2). Therefore, performing the position ordering, the single scattering DDCS, Eq. (4.12), becomes

$$\frac{d^2\sigma(1)}{d\Omega dE_f} = \lim_{\varepsilon\to 0} \frac{m_N^2}{(2\pi\hbar)^3}\frac{1}{\hbar^2}\frac{k_f}{k_i}$$

$$\times \frac{d}{dt}\sum_{\vec{k}_1,\vec{k}_2}\int_{-\infty}^{t} dt_1 \int_{-\infty}^{t} dt_2 g(\vec{k}_1)g(\vec{k}_2)\langle \vec{k}_i | e^{i\vec{k}_1\cdot\vec{r}_N(t_1)} | \vec{k}_f\rangle$$

$$\times \langle \vec{k}_f | e^{i\vec{k}_2\cdot\vec{r}_N(t_2)} | \vec{k}_i\rangle \langle n_{\vec{k}_1}(t_1) n_{\vec{k}_2}(t_2)\rangle e^{\varepsilon(t_1+t_2)} \quad (4.18)$$

The neutron matrix elements appearing in Eq. (4.18) are

$$\langle \vec{k}_i | e^{i\vec{k}_1\cdot\vec{r}_N(t_1)} | \vec{k}_f\rangle = e^{i\omega t_1}\delta_{\vec{k},\vec{k}_1} \quad (4.19)$$

$$\langle \vec{k}_f | e^{i\vec{k}_2\,\vec{r}_N(t_2)} | \vec{k}_i\rangle = e^{-i\omega t_2}\delta_{-\vec{k},\vec{k}_2} \quad (4.20)$$

where $\hbar\omega = E_i - E_f$ is the neutron energy loss and $\hbar\vec{k} = \hbar(\vec{k}_i - \vec{k}_f)$ is the neutron momentum transfer. Therefore, Eq. (4.18) becomes

$$\frac{d^2\sigma(1)}{d\Omega dE_f} = \lim_{\varepsilon\to 0} \frac{m_N^2}{(2\pi\hbar)^3}\frac{1}{\hbar^2}\frac{k_f}{k_i}\,|g(\vec{k})|^2$$

$$\times \frac{d}{dt}\int_{-\infty}^{t} dt_1 \int_{-\infty}^{t} dt_2\, e^{i(\omega-i\varepsilon)t_1} e^{-i(\omega+i\varepsilon)t_2} \langle n_{\vec{k}}(t_1) n_{-\vec{k}}(t_2)\rangle \quad (4.21)$$

The target average, $\langle n_{\vec{k}}(t_1) n_{-\vec{k}}(t_2)\rangle$, can be expressed in terms of the dynamic structure factor, $S(\vec{k},\omega)$, in the form

$$\langle n_{\vec{k}}(t_1) n_{-\vec{k}}(t_2) \rangle = \int d\omega' \, e^{i\omega'(t_2 - t_1)} S(\vec{k},\omega') \tag{4.22}$$

or

$$S(\vec{k},\omega') = \frac{1}{2\pi} \int d\tau \, e^{-i\omega'\tau} \langle n_{\vec{k}}(0) n_{-\vec{k}}(\tau) \rangle \tag{4.23}$$

where $\tau = t_2 - t_1$, and the fact that $\langle n_{\vec{k}}(t_1) n_{-\vec{k}}(t_2) \rangle$ is invariant under a time translation has been used. Inserting Eq. (4.22) into (4.21), the time integrations are

$$\lim_{\varepsilon \to 0} \frac{d}{dt} \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t} dt_2 \, e^{i(\omega-\omega'-i\varepsilon)t_1} \, e^{-i(\omega-\omega'+i\varepsilon)t_2}$$

$$= \lim_{\varepsilon \to 0} \frac{d}{dt} \left[\frac{e^{2\varepsilon t}}{(\omega-\omega')^2+\varepsilon^2} \right] = \lim_{\varepsilon \to 0} \left[\frac{2\varepsilon e^{2\varepsilon t}}{(\omega-\omega')^2+\varepsilon^2} \right]$$

$$\simeq \lim_{\varepsilon \to 0} \left[\frac{2\varepsilon}{(\omega-\omega')^2+\varepsilon^2} \right] = 2\pi\delta(\omega-\omega') \tag{4.24}$$

where condition (3.24) has been utilized, and

$$\delta(x) = \frac{1}{\pi} \lim_{\varepsilon \to 0} \frac{\varepsilon}{x^2+\varepsilon^2} \tag{4.25}$$

is the familiar Dirac delta function. From Eqs. (4.22) and (4.24), the first Born DDCS, Eq. (4.21), can be written

$$\frac{d^2\sigma(1)}{d\Omega dE_f} = \frac{m_N^2}{4\pi^2\hbar^5}\frac{k_f}{k_i}\,|g(\vec{k})|^2 S(\vec{k},\omega) \tag{4.26}$$

which is in the well-known form first given by Van Hove.[3]

For magnetic scattering, the interaction, ϕ, depends not only on the relative neutron - j^{th} target electron separation, $\vec{r} = \vec{r}_N - \vec{r}_j$, but also on the neutron's magnetic moment, $\vec{\mu}_N$, and the j^{th} target electron's magnetic moment, $\vec{\mu}_j$. Therefore, the interaction can be written

$$\phi(t) = \sum_j \phi[\vec{r}(t),\vec{\mu}_N,\vec{\mu}_j(t)]$$

$$= \sum_j \sum_{\vec{k}_1} \hat{\phi}[\vec{k}_1,\vec{\mu}_N,\vec{\mu}_j(t)]\; e^{i\vec{k}_1\cdot\vec{r}_N(t)}\; e^{-i\vec{k}_1\cdot\vec{r}_j(t)} \tag{4.27}$$

where

$$\hat{\phi}[\vec{k}_1,\vec{\mu}_N,\vec{\mu}_j(t)] = \int d^3r\; e^{-i\vec{k}_1\cdot\vec{r}}\; \phi[\vec{r},\vec{\mu}_N,\vec{\mu}_j(t)] \tag{4.28}$$

Because of the simple form of the magnetic dipole interaction, ϕ, (see Chapter VII), its Fourier transform, $\hat{\phi}$, can be written

$$\hat{\phi}[\vec{k}_1,\vec{\mu}_N,\vec{\mu}_j(t)] = \vec{a}(\vec{\mu}_N)\cdot\vec{b}[\vec{k}_1,\vec{\mu}_j(t)] = a^\alpha b^\alpha \tag{4.29}$$

where $\alpha = x,y,z$ refer to rectangular coordinates, and the Einstein con-

vention of summing over repeated indices is adopted. Therefore, the magnetic dipole interaction may be written as

$$\phi(t) = \sum_{\vec{k}_1} A^{\alpha}(\vec{k}_1,t)B^{\alpha}(\vec{k}_1,t) \tag{4.30}$$

where, from Eqs. (4.27) and (4.29)

$$A^{\alpha}(\vec{k}_1,t) = e^{i\vec{k}_1\cdot\vec{r}_N(t)} \; a^{\alpha}(\vec{\mu}_N) \tag{4.31}$$

and

$$B^{\alpha}(\vec{k}_1,t) = \sum_{j} e^{-i\vec{k}_1\cdot\vec{r}_j(t)} \; b^{\alpha}[\vec{k}_1,\vec{\mu}_j(t)] \tag{4.32}$$

The A operator contains only neutron operators and the B operator contains only target operators; thus, the interaction, ϕ, is in the desired form, Eq. (4.2).

The initial and final neutron states, as they appear in the cross section, Eq. (4.1), must now be written as

$$|\vec{k}_{i,f}\rangle|\sigma_{i,f}\rangle,$$

i.e., as a direct product of neutron spatial and spin states. Thus, making this identification in Eq. (4.12), using Eqs. (4.31) and (4.32) in Eq. (4.12) and following the same analysis as presented through Eq. (4.26), the first Born DDCS for magnetic scattering is given by

$$\frac{d^2\sigma(1)}{d\Omega dE_f} = \frac{m_N^2}{4\pi^2\hbar^5}\frac{k_f}{k_i} M_N^{\alpha\beta} S^{\alpha\beta}(\vec{k},\omega) \tag{4.33}$$

where $M_N^{\alpha\beta}$ stands for the neutron spin matrix elements

$$M_N^{\alpha\beta} = \langle\sigma_i|a^\alpha(\vec{\mu}_N)|\sigma_f\rangle\langle\sigma_f|a^\beta(\vec{\mu}_N)|\sigma_i\rangle \tag{4.34}$$

and the dynamic structure factor is now

$$S^{\alpha\beta}(\vec{k},\omega) = \frac{1}{2\pi}\int d\tau\ e^{-i\omega\tau}\langle B^\alpha(\vec{k},0)B^\beta(-\vec{k},\tau)\rangle \tag{4.35}$$

The target average is now over both target spatial and spin states.

Double Scattering

Retaining only the middle term in Δ_4, Eq. (4.10), in the DDCS, Eq. (4.11), results in the double scattering DDCS

$$\begin{aligned}\frac{d^2\sigma(2)}{d\Omega dE_f} = &\lim_{\varepsilon\to 0}\frac{m_N^2}{(2\pi\hbar)^3}\frac{1}{\hbar^4}\frac{k_f}{k_i}\\ &\times P_{12}\frac{d}{dt}\langle\vec{k}_{f_2}|\langle\vec{k}_{i_1}|\int_{-\infty}^{t}dt_1\int_{-\infty}^{t_1}dt_2\int_{-\infty}^{t}dt_3\int_{-\infty}^{t_3}dt_4\\ &\times A_1(t_2)A_1(t_1)A_2(t_3)A_2(t_4)|\vec{k}_{i_2}\rangle|\vec{k}_{f_i}\rangle\\ &\times\langle B(t_2)B(t_1)B(t_3)B(t_4)\rangle\, e^{\varepsilon(t_1+t_2+t_3+t_4)}\end{aligned} \tag{4.36}$$

which contains the average over initial target states of four B operators. If double scattering is to be a small effect, then to a good approximation, the four B operator correlation function appearing in Eq. (4.36) can be decomposed into products of pair correlation functions. To give a plausibility argument in support of this supposition, consider an interaction, ϕ, that depends on distance only; thus B(t) is given by Eq. (4.17). Each operator B is evaluated at a space-time point of the target. Now, if double scattering is to be a small effect, then one would expect the two scattering events to be well separated in space within the target, i.e., the two events are uncorrelated for most targets. In this regard, the double scattering can be considered as a convolution of two single scatterings. For this type of double scattering, two of the B operators in $\langle BBBB \rangle$ are required to be evaluated at space-time points that are well separated from the space-time points at which the other two B operators are evaluated. Therefore, unless a particular target has a very long correlation range and relaxation time, the four B operator correlation function can be decomposed into products of pair correlations,

$$\begin{aligned} \langle B(t_2)B(t_1)B(t_3)B(t_4)\rangle = {} & \langle B(t_2)B(t_1)\rangle\langle B(t_3)B(t_4)\rangle \\ & + \langle B(t_2)B(t_3)\rangle\langle B(t_1)B(t_4)\rangle \\ & + \langle B(t_2)B(t_4)\rangle\langle B(t_1)B(t_3)\rangle \end{aligned} \tag{4.37}$$

For target systems near their critical points, correlation ranges and relaxation times can become very long. Therefore, from the above discussion, one might infer that the decomposition given by Eq. (4.37) would not be applicable. However, this is not the case and with a slight modification, the above decomposition is still applicable. This point will be discussed in Chapter VI where critical nuclear double scattering is treated.

Of the three terms on the right-hand-side of Eq. (4.37), only the third term is found to contribute significantly to double scattering and will be the only term retained. The first two terms are treated in Chapter VI within the context of critical nuclear scattering.

Retaining only the third term in Eq. (4.37), the double scattering DDCS, Eq. (4.36), becomes

$$\frac{d^2\sigma(2)}{d\Omega dE_f} = \lim_{\varepsilon\to 0} \frac{m_N^2}{(2\pi\hbar)^3} \frac{1}{\hbar^4} \frac{k_f}{k_i}$$

$$\times P_{12} \frac{d}{dt} \langle \vec{k}_{f_2} | \langle \vec{k}_{i_1} | \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t} dt_3 \int_{-\infty}^{t_3} dt_4$$

$$\times A_1(t_2)A_1(t_1)A_2(t_3)A_2(t_4) | \vec{k}_{i_2} \rangle | \vec{k}_{f_1} \rangle$$

$$\times \langle B(t_2)B(t_4) \rangle \langle B(t_1)B(t_3) \rangle \, e^{\varepsilon(t_1 + t_2 + t_3 + t_4)} \qquad (4.38)$$

Considering an interaction that depends on distance only, ϕ is given

by Eq. (4.15) and the A and B operators are given by Eqs. (4.16) and (4.17). Thus, after performing the position ordering, the double scattering DDCS, Eq. (4.38), becomes

$$\frac{d^2\sigma(2)}{d\Omega dE_f} = \lim_{\varepsilon\to 0} \frac{m_N^2}{(2\pi\hbar)^3} \frac{1}{\hbar^4} \frac{k_f}{k_i}$$

$$\times \frac{d}{dt} \sum_{\vec{k}_1,\vec{k}_2,\vec{k}_3,\vec{k}_4} \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t} dt_3 \int_{-\infty}^{t_3} dt_4$$

$$\times g(\vec{k}_1)g(\vec{k}_2)g(\vec{k}_3)g(\vec{k}_4) \langle \vec{k}_i | e^{i\vec{k}_2\cdot\vec{r}_N(t_2)} e^{i\vec{k}_1\cdot\vec{r}_N(t_1)} | \vec{k}_f \rangle$$

$$\times \langle \vec{k}_f | e^{i\vec{k}_3\cdot\vec{r}_N(t_3)} e^{i\vec{k}_4\cdot\vec{r}_N(t_4)} | \vec{k}_i \rangle \langle n_{\vec{k}_2}(t_2) n_{\vec{k}_4}(t_4) \rangle$$

$$\times \langle n_{\vec{k}_1}(t_1) n_{\vec{k}_3}(t_3) \rangle e^{\varepsilon(t_1 + t_2 + t_3 + t_4)} \qquad (4.39)$$

The neutron matrix elements appearing in Eq. (4.39) are expressed in terms of the intermediate neutron states with energies E_a and E_b and wave vectors $\vec{k}_a$ and $\vec{k}_b$ in the following form

$$\langle \vec{k}_i | e^{i\vec{k}_2\cdot\vec{r}_N(t_2)} e^{i\vec{k}_1\cdot\vec{r}_N(t_1)} | \vec{k}_f \rangle = \sum_{\vec{k}_a} e^{i(E_i - E_a)t_2/\hbar} e^{i(E_a - E_f)t_1/\hbar}$$

$$\times \delta_{\vec{k}_i,\vec{k}_2 + \vec{k}_a} \delta_{\vec{k}_a,\vec{k}_1 + \vec{k}_f} \qquad (4.40)$$

$$\langle \vec{k}_f | e^{i\vec{k}_3\cdot\vec{r}_N(t_3)} e^{i\vec{k}_4\cdot\vec{r}_N(t_4)} | \vec{k}_i \rangle = \sum_{\vec{k}_b} e^{i(E_f - E_b)t_3/\hbar} e^{i(E_b - E_i)t_4/\hbar}$$

$$\times \delta_{\vec{k}_f,\vec{k}_3 + \vec{k}_b} \delta_{\vec{k}_b,\vec{k}_4 + \vec{k}_i} \tag{4.41}$$

Following Van Hove, a dynamic structure factor, $S(\vec{k}_2,\vec{k}_4,\omega')$, is defined[10]

$$\langle n_{\vec{k}_2}(t_2) n_{\vec{k}_4}(t_4) \rangle \equiv \int d\omega' \, e^{i\omega'(t_4 - t_2)} S(\vec{k}_2,\vec{k}_4,\omega') \tag{4.42}$$

or

$$S(\vec{k}_2,\vec{k}_4,\omega') = \frac{1}{2\pi} \int d\tau \; e^{-i\omega'\tau} \langle n_{\vec{k}_2}(0) n_{\vec{k}_4}(\tau) \rangle \tag{4.43}$$

where $\tau = t_4 - t_2$. From Eqs. (4.40), (4.41) and (4.42), the double scattering DDCS, Eq. (4.39), becomes

$$\frac{d^2\sigma(2)}{d\Omega dE_f} = \lim_{\varepsilon\to 0} \frac{m_N^2}{(2\pi\hbar)^3} \frac{1}{\hbar^4} \frac{k_f}{k_i}$$

$$\times \frac{d}{dt} \sum_{k_a,k_b} \int d\omega' \int d\omega'' \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t} dt_3 \int_{-\infty}^{t_3} dt_4$$

$$\times g(\vec{k}_a - \vec{k}_f) g(\vec{k}_i - \vec{k}_a) g(\vec{k}_f - \vec{k}_b) g(\vec{k}_b - \vec{k}_i)$$

$$\times e^{i(E_a - E_f - \hbar\omega'')t_1/\hbar} e^{i(E_i - E_a - \hbar\omega')t_2/\hbar}$$

$$\times e^{i(E_f - E_b + \hbar\omega'')t_3/\hbar} e^{i(E_b - E_i + \hbar\omega')t_4/\hbar}$$

$$\times e^{\varepsilon(t_1 + t_2 + t_3 + t_4)} S(\vec{k}_i - \vec{k}_a, \vec{k}_b - \vec{k}_i, \omega')$$

$$\times S(\vec{k}_a - \vec{k}_f, \vec{k}_f - \vec{k}_b, \omega'') \qquad (4.44)$$

The time integrations in Eq. (4.44) yield

$$\frac{d}{dt} \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t} dt_3 \int_{-\infty}^{t_3} dt_4 \, e^{i(E_a - E_f - \hbar\omega'')t_1/\hbar}$$

$$\times e^{i(E_i - E_a - \hbar\omega')t_2/\hbar} e^{i(E_f - E_b + \hbar\omega'')t_3/\hbar}$$

$$\times e^{i(E_b - E_i + \hbar\omega')t_4/\hbar} e^{\varepsilon(t_1 + t_2 + t_3 + t_4)}$$

$$= 2\pi\hbar^2 \left[(E_a - E_i + \hbar\omega' + i\hbar\varepsilon)(E_b - E_i + \hbar\omega' - i\hbar\varepsilon)\right]^{-1}$$

$$\times \delta(\omega - \omega' - \omega'') \qquad (4.45)$$

where condition (3.24) has been utilized and $\delta(\omega - \omega' - \omega'')$ has

replaced

$$\frac{1}{\pi}\frac{\varepsilon}{(\omega - \omega' - \omega'')^2 + \varepsilon^2}$$

in anticipation of the limit $\varepsilon \to 0$. Inserting Eq. (4.45) into Eq. (4.44) and letting the sums over $\vec{k}_a$ and $\vec{k}_b$ go over to integrals,

$$\sum_{\vec{k}_a} \to \frac{1}{(2\pi)^3}\int d^3k_a$$

i.e., letting the quantization volume go to infinity, the double scattering DDCS becomes

$$\frac{d^2\sigma(2)}{d\Omega dE_f} = \lim_{\varepsilon\to 0} \frac{4m_N^4}{(2\pi\hbar)^8}\frac{1}{\hbar}\frac{k_f}{k_i}\int d^3k_a \int d^3k_b \int d\omega'$$

$$\times g(\vec{k}_a - \vec{k}_f)g(\vec{k}_i - \vec{k}_a)g(\vec{k}_f - \vec{k}_b)g(\vec{k}_b - \vec{k}_i)$$

$$\times S(\vec{k}_i - \vec{k}_a, \vec{k}_b - \vec{k}_i, \omega')S(\vec{k}_a - \vec{k}_f, \vec{k}_f - \vec{k}_b, \omega - \omega')$$

$$\times (k_a^2 - k_i^2 + \frac{2m_N\omega'}{\hbar} + i\varepsilon)^{-1}\ (k_b^2 - k_i^2 + \frac{2m_N\omega'}{\hbar} - i\varepsilon)^{-1} \qquad (4.46)$$

For magnetic scattering, the interaction ϕ is given by Eq. (4.30) and the A and B operators are given by Eqs. (4.31) and (4.32). Thus,

the corresponding double scattering DDCS for magnetic scattering is given by

$$\frac{d^2\sigma(2)}{d\Omega dE_f} = \lim_{\varepsilon\to 0} \frac{4m_N^4}{(2\pi\hbar)^8} \frac{1}{\hbar} \frac{k_f}{k_i} \int d^3k_a \int d^3k_b \int d\omega'$$

$$\times M_N^{\alpha\beta\gamma\delta}\, S^{\alpha\delta}(\vec{k}_i - \vec{k}_a,\ \vec{k}_b - \vec{k}_i, \omega') S^{\beta\gamma}(\vec{k}_a - \vec{k}_f,\ \vec{k}_f - \vec{k}_b, \omega - \omega')$$

$$\times \left(k_a^2 - k_i^2 + \frac{2m_N\omega'}{\hbar} + i\varepsilon\right)^{-1} \left(k_b^2 - k_i^2 + \frac{2m_N\omega'}{\hbar} - i\varepsilon\right)^{-1} \qquad (4.47)$$

where $M_N^{\alpha\beta\gamma\delta}$ stands for the neutron spin matrix elements

$$M_N^{\alpha\beta\gamma\delta} = \langle\sigma_i|a^\alpha(\vec{\mu}_N)a^\beta(\vec{\mu}_N)|\sigma_f\rangle\langle\sigma_f|a^\gamma(\vec{\mu}_N)a^\delta(\vec{\mu}_N)|\sigma_i\rangle \qquad (4.48)$$

and the dynamic structure factor, $S^{\alpha\delta}(\vec{k}_2,\vec{k}_4,\omega')$, is now given by

$$S^{\alpha\delta}(\vec{k}_2,\vec{k}_4,\omega') = \frac{1}{2\pi}\int d\tau\ e^{-i\omega'\tau}\langle B^\alpha(\vec{k}_2,0)B^\delta(\vec{k}_4,\tau)\rangle \qquad (4.49)$$

The double scattering DDCS as given by Eq. (4.46) or Eq. (4.47) is expressed in terms of products of two dynamic structure factors and thus can be considered as a convolution of two single scatterings. The remainder of this treatment will deal exclusively with the critical (nuclear and magnetic) scattering of thermal-neutrons from targets at temperatures slightly above their critical temperature.

CHAPTER V

CRITICAL NUCLEAR SINGLE SCATTERING FROM LIQUIDS AND THE ORNSTEIN-ZERNIKE CORRELATION FUNCTION

In this chapter, the single scattering cross section for critical nuclear scattering of thermal-neutrons from a liquid is obtained. The classical Ornstein-Zernike correlation function, which is used in obtaining the single scattering cross section, is also obtained using the methods of Klein and Tisza[5] and Ornstein and Zernike.[4]

Critical Nuclear Single Scattering

In considering the nuclear scattering of thermal-neutrons, the actual interaction between the neutron and the nuclei of the target is well approximated by its S-wave approximation, i.e., the Fermi-pseudo potential,

$$\phi = \sum_j \phi(\vec{r}) = \frac{2\pi\hbar^2 a}{m_N} \sum_j \delta(\vec{r}_N - \vec{r}_j) \tag{5.1}$$

where a is the nuclear S-wave scattering length and is assumed to be identical for all nuclei and spin independent, and $\vec{r} = \vec{r}_N - \vec{r}_j$, where $\vec{r}_N$ is the position vector of the neutron and $\vec{r}_j$ is the position vector of the j^{th} target nucleus. The Fourier transform of $\phi(\vec{r})$, Eq. (4.14), is thus given by

$$g = \frac{2\pi\hbar^2 a}{m_N} \tag{5.2}$$

and the single scattering DDCS becomes, from Eq. (4.26),

$$\frac{d^2\sigma(1)}{d\Omega dE_f} = \frac{a^2}{\hbar} \frac{k_f}{k_i} S(\vec{k},\omega) \tag{5.3}$$

where the dynamic structure factor, $S(\vec{k},\omega)$, is given by Eq. (4.23).

The single scattering DDCS, Eq. (5.3), can be greatly simplified for critical scattering, i.e., for neutrons scattering from a liquid near its critical point. As will be shown later, density fluctuations become very large in a liquid near its critical point, and it is these density fluctuations that scatter neutrons. Also, these density fluctuations have a very long relaxation time compared to the time it takes a neutron to traverse an effective scattering volume. Therefore, a neutron will "see" a static picture of the liquid and to a good approximation

$$n_{\vec{k}}(\tau) \tilde{=} n_{\vec{k}}(0) \tag{5.4}$$

Therefore, the dynamic structure factor, Eq. (4.23), becomes

$$S(\vec{k},\omega) \tilde{=} \frac{1}{2\pi} \int d\tau\ e^{-i\omega\tau} \langle n_{\vec{k}}(0) n_{-\vec{k}}(0) \rangle = \langle n_{\vec{k}} n_{-\vec{k}} \rangle \delta(\omega) \tag{5.5}$$

Writing k_f in terms of $\omega = \frac{1}{\hbar}(E_i - E_f)$,

$$k_f = \left[k_i^2 - \frac{2m_N\omega}{\hbar} \right]^{\frac{1}{2}} \tag{5.6}$$

the single scattering DDCS, Eq. (5.3), becomes

$$\frac{d^2\sigma(1)}{d\Omega dE_f} = \frac{a^2}{\hbar} \langle n_{\vec{k}} n_{-\vec{k}} \rangle \delta(\omega) \tag{5.7}$$

where the mathematical identity,

$$f(x)\delta(x) = f(0)\delta(x) \tag{5.8}$$

has been used. The approximation given by Eq. (5.4) is called the quasielastic approximation (QA) since it requires $k_i = k_f$, i.e., the neutron's initial energy to equal its final energy, yet still allows the target to make transitions to different energy eigenstates as implied in $\langle n_{\vec{k}} n_{-\vec{k}} \rangle$. Physically, the QA is valid for those scattering systems where the energy loss, $\hbar\omega$, by the probe is small compared to the probe's initial energy; thus, to a good approximation, this loss can be neglected for probe considerations, i.e., $k_i \cong k_f$. However, this energy, $\hbar\omega$, when transferred to the target may be large compared to the separation between target energy levels and, thus, cannot be totally neglected for target considerations. It is accounted for by allowing the target to make transitions to different energy eigenstates.

Integrating Eq. (5.7) over final neutron energies, the single scattering DCS is obtained,

$$\frac{d\sigma}{d\Omega}^{(1)} = a^2 \langle n_{\vec{k}} n_{-\vec{k}} \rangle \tag{5.9}$$

From Eq. (4.17), the target average in Eq. (5.9) can be written as

$$\langle n_{\vec{k}} n_{-\vec{k}} \rangle = \langle \sum_j e^{-i\vec{k}\cdot\vec{r}_j} \sum_\ell e^{i\vec{k}\cdot\vec{r}_\ell} \rangle$$

$$= \int d^3r_1 \int d^3r_2 \, e^{-i\vec{k}\cdot(\vec{r}_1 - \vec{r}_2)} \langle n(\vec{r}_1) n(\vec{r}_2) \rangle \tag{5.10}$$

where

$$n(\vec{r}) = \sum_j \delta(\vec{r} - \vec{r}_j) \tag{5.11}$$

is the particle number density operator.

In the asymptotic limit $|\vec{r}_1 - \vec{r}_2| \to \infty$, the density operators $n(\vec{r}_1)$ and $n(\vec{r}_2)$ become uncorrelated, i.e., $\langle n(\vec{r}_1) n(\vec{r}_2) \rangle \to \langle n(\vec{r}_1) \rangle \langle n(\vec{r}_2) \rangle$ and the scattering becomes purely elastic. Therefore, writing

$$\langle n(\vec{r}_1) n(\vec{r}_2) \rangle = \{ \langle n(\vec{r}_1) n(\vec{r}_2) \rangle - \langle n(\vec{r}_1) \rangle \langle n(\vec{r}_2) \rangle \} + \langle n(\vec{r}_1) \rangle \langle n(\vec{r}_2) \rangle \tag{5.12}$$

the single scattering DCS, Eq. (5.9), can be separated into a purely elastic part and an inelastic part,

$$\frac{d\sigma}{d\Omega}^{(1)} = \left(\frac{d\sigma}{d\Omega} \right)_e^{(1)} + \left(\frac{d\sigma}{d\Omega} \right)_i^{(1)} \tag{5.13}$$

where

$$\left(\frac{d\sigma}{d\Omega}\right)_e^{(1)} = a^2 \int d^3r_1 \int d^3r_2 \; e^{-i\vec{k}\cdot(\vec{r}_1 - \vec{r}_2)} \langle n(\vec{r}_1)\rangle \langle n(\vec{r}_2)\rangle \tag{5.14}$$

$$\left(\frac{d\sigma}{d\Omega}\right)_i^{(1)} = a^2 \int d^3r_1 \int d^3r_2 \; e^{-i\vec{k}\cdot(\vec{r}_1 - \vec{r}_2)} \Gamma(\vec{r}_1,\vec{r}_2) \tag{5.15}$$

and

$$\Gamma(\vec{r}_1,\vec{r}_2) = \langle n(\vec{r}_1)n(\vec{r}_2)\rangle - \langle n(\vec{r}_1)\rangle\langle n(\vec{r}_2)\rangle \tag{5.16}$$

is the familiar density-density correlation function. The elastic DCS is non-negligible only in the forward ($\vec{k} = 0$) scattering direction for macroscopic targets. For a target of constant mean number density, ρ, at every point r,

$$\langle n(\vec{r})\rangle = \rho \qquad (\vec{r} \text{ inside the target}) \tag{5.17}$$

and

$$\left(\frac{d\sigma}{d\Omega}\right)_e^{(1)} \cong (2\pi)^3 \, a^2 N\rho\,\delta(\vec{k}) \tag{5.18}$$

The elastic DCS will now be dropped from further considerations.

As can be seen from Eq. (5.15), in general, the single scat-

tering inelastic DCS, as given in the QA, depends on how the correlation of target particle densities at two points in the liquid fluctuates from its mean value. Therefore, by measuring the single scattering inelastic DCS, the static structure of the liquid can be determined. For liquid systems where the relaxation times of these density fluctuations are comparable to or greater than the time it takes a probe to traverse an effective scattering volume, i.e., where the QA cannot be made, then, as shown by Van Hove,[3] measurement of the single scattering DDCS will determine the dynamic structure of the liquid. The single scattering inelastic DCS, Eq. (5.15), will now be determined for the case where the liquid target is near its critical point.

The critical point of a liquid is defined by a point on a three dimensional pressure-density-temperature (P-ρ-T) surface with coordinates P_c, ρ_c and T_c which are, respectively, the critical pressure, the critical density and the critical temperature. The P-ρ-T surface is determined by the equation of state, $f(P,\rho,T) = 0$. If a gas is isothermally compressed at a temperature $T > T_c$, it will not undergo a phase transition, i.e., it will remain a gas. If compressed at a temperature $T < T_c$, the gas will undergo a "first order" phase transition, i.e., it will enter a two-phase region on the P-ρ-T surface in which it can coexist in equilibrium in both its gaseous and liquid phase. However, as the gas is compressed at $T = T_c$, it will undergo a "second order" phase transition at the critical point in which it continuously goes from a gas to a liquid at that point, i.e., there is no two-phase region.

Consider a system held fixed at its mean critical density,[11] $\rho = \rho_c$, (the system is said to be on the critical isochore) and brought to its critical point by lowering its temperature towards T_c, i.e., $T \to T_c^+$. As the system approaches T_c, the fluctuations in its density will become very large since it is approaching its liquid phase and the atoms of the system are spontaneously forming large clusters and breaking up. Thus, the single scattering inelastic DCS, Eq. (5.15), will become very large. This can be seen in a qualitative way by adopting as the equation of state for the system the van der Waals equation. It readily follows from this equation that on the critical isochore, the isothermal compressibility, K_T, is given by[12]

$$K_T \equiv \frac{1}{V}\left(\frac{\partial V}{\partial P}\right)_T = \frac{A}{|T - T_c|} \tag{5.19}$$

where V is the volume of the target and A is a constant. Using standard methods from statistical physics, it can also be shown that[13]

$$\int d^3r_1 \int d^3r_2 \; \Gamma(\vec{r}_1, \vec{r}_2) = \frac{N^2}{V} k_B T K_T \tag{5.20}$$

where N is the number of atoms in the system and k_B is Boltzmann's constant. Therefore, from Eqs. (5.15), (5.19) and (5.20) it follows that

$$\left(\frac{d\sigma}{d\Omega}\right)_i^{(1)}\Bigg|_{\vec{k} = 0} \to \infty \qquad \text{as } T \to T_c \tag{5.21}$$

i.e., the forward scattering becomes huge near the critical point. This phenomena in light scattering is known as critical opalesence.

Assuming translational invariance for the liquid target, i.e., neglecting boundary effects, one can write

$$\int d^3r_1 \int d^3r_2 \, \Gamma(\vec{r}_1,\vec{r}_2) = V \int d^3r \, \Gamma(\vec{r}) \tag{5.22}$$

where $\vec{r} = \vec{r}_2 - \vec{r}_1$. The density-density correlation function, $\Gamma(\vec{r})$, can, in general, be written as

$$\Gamma(\vec{r}) = \rho\delta(\vec{r}) + \rho^2 g(\vec{r}) \tag{5.23}$$

where $\rho\delta(\vec{r})$ is the self-correlation function, arising from the correlation of a target particle with itself. The function $g(\vec{r})$ is the pair correlation function and arises from the correlation between two different target particles. Therefore, from Eqs. (5.22) and (5.23), Eq. (5.20) becomes

$$1 + \rho \int d^3r \, g(\vec{r}) = \rho k_B T K_T \tag{5.24}$$

As $T \to T_c$, the integral in Eq. (5.24) must diverge; therefore $g(\vec{r})$ must be a long-ranged function that falls off at least as slow as r^{-3} when $T = T_c$. That the pair correlation function be a long-ranged function as $T \to T_c$ is an obvious physical necessity in order to produce the large density fluctuations observed near the critical point.

The single scattering inelastic DCS, Eq. (5.15), becomes, from Eqs. (5.22) and (5.23)

$$\left(\frac{d\sigma}{d\Omega}\right)_i^{(1)} = Na^2 \left[1 + \rho \int d^3r\, e^{i\vec{k}\cdot\vec{r}} g(\vec{r})\right] \tag{5.25}$$

The self-correlation term, $\rho\delta(\vec{r}_1)$, in Eq. (5.23) gives rise to the isotropic scattering term Na^2 in the DCS, Eq. (5.25). Dropping this term from further considerations, the angular-dependent single scattering inelastic DCS is given by

$$\left(\frac{d\sigma}{d\Omega}\right)_i^{(1)} = N\rho a^2 \int d^3r\, e^{i\vec{k}\cdot\vec{r}} g(\vec{r}) \tag{5.26}$$

For small angle (small k) scattering, g(r) need only be determined in the asymptotic limit of large r, and in this limit, a classical determination is adequate. The derivation of $g(\vec{r})$ for a liquid near its critical point was first given classically by Ornstein and Zernike and later improved upon by Klein and Tisza. A brief review of their methods will now be given.

The Classical Ornstein-Zernike Correlation Function

Following Klein and Tisza, consider a system with fixed volume V divided up into M identical cells. Now consider one arbitrary extensive variable, y, (other than V) of the system, holding all other variables fixed. The value of this variable in the k^{th} cell is denoted y_k, and a statistical state of the system is given by specifying the value of y in all cells, i.e., by the set of numbers $\{y_k\}$. Klein and

Tisza showed that the free energy, F, of the system is adequately given by the quadratic form

$$F = \frac{1}{2} C' \sum_{k,\ell=1}^{M} a_{k\ell} z_k z_\ell \tag{5.27}$$

where C' is a constant. The constant $a_{k\ell}$ is a measure of the interaction between the k^{th} cell and the ℓ^{th} cell and is symmetric, i.e., $a_{k\ell} = a_{\ell k}$. The variable z_k is the fluctuation of y_k about its mean value, y,

$$z_k = y_k - y \tag{5.28}$$

The probability, P, of finding the system in state $\{y_k\}$ is given by

$$P = C\, e^{-\frac{\theta}{2} \sum_{k,\ell} a_{k\ell} z_k z_\ell} \tag{5.29}$$

where C is a constant and

$$\theta = (k_B T)^{-1} \tag{5.30}$$

Considering $a_{k\ell}$ to be the elements of a matrix A, then, as shown in Appendix II, the correlation between z_k and z_ℓ is given by

$$\langle z_k z_\ell \rangle = \theta^{-1} (A^{-1})_{k\ell} \tag{5.31}$$

where A^{-1} is the inverse of A. Thus,

$$\langle z_k z_\ell \rangle = \theta^{-1} \frac{A_{k\ell}}{A} \tag{5.32}$$

where $A_{k\ell}$ is the cofactor of A (with appropriate sign) and $A = \det A$. The correlation function, $g_{k\ell}$, is defined as

$$g_{k\ell} = \frac{\langle z_k z_\ell \rangle}{[\langle z_k^2 \rangle \langle z_\ell^2 \rangle]^{\frac{1}{2}}} \tag{5.33}$$

For a translationally invariant system, from Eq. (5.32),

$$A_{kk} = A_{\ell\ell} \tag{5.34}$$

Therefore, the correlation function, Eq. (5.33), becomes, from Eqs. (5.32) and (5.34)

$$g_{k\ell} = \frac{A_{k\ell}}{A_{\ell\ell}} \tag{5.35}$$

Considering only the case where $k \neq \ell$, and defining the direct correlation function, $c_{k\ell}$,

$$c_{k\ell} = - \frac{a_{k\ell}}{a_{kk}} \tag{5.36}$$

then

$$\sum_{m\neq k,\ell} c_{km}g_{m\ell} = g_{k\ell} - c_{k\ell} \qquad (k\neq\ell) \tag{5.37}$$

or

$$g_{k\ell} = c_{k\ell} + \sum_{m\neq k,\ell} c_{km}g_{m\ell} \qquad (k\neq\ell) \tag{5.38}$$

For a translationally invariant system,

$$\langle z_k z_\ell \rangle = \langle z_0 z_{\ell-k} \rangle = \langle z_0 z_p \rangle \tag{5.39}$$

therefore,

$$g_k = c_k + \sum_{m\neq k} c_{km}g_m \tag{5.40}$$

Choosing $y_k = N_k$, from Eq. (5.28)

$$z_k = v_o(\rho_k - \rho) \tag{5.41}$$

where v_o is the volume of a cell, ρ_k is the particle number density in the k^{th} cell and ρ is the mean particle number density. Since the primary interest will be in obtaining g_k for large k, the volume of each cell can be taken so small that each cell contains, on the average,

only one particle and Eq. (5.40) can be written

$$g(r) \cong c(r) + \rho \int d^3r' \, c(r' - r)g(r') \tag{5.42}$$

which is the well-known integral equation of Ornstein and Zernike.

Taking the Fourier transform of Eq. (5.42) yields

$$\hat{g}(k) = \hat{c}(k) + \rho\hat{c}(k)\hat{g}(k) \tag{5.43}$$

where

$$\hat{g}(k) = \int d^3r \, g(r) \, e^{-i\vec{k}\cdot\vec{r}} \tag{5.44}$$

$$\hat{c}(k) = \int d^3r \, c(r) \, e^{-i\vec{k}\cdot\vec{r}} \tag{5.45}$$

and the convolution theorem from Fourier analysis has been utilized.[14] Ornstein and Zernike assumed that c(r), the direct correlation function, is a short-ranged function, implying that $\hat{c}(k)$ is a long-ranged function. Thus, if $\hat{c}(k)$ can be expanded in a Taylor's series about k = 0, then it would be sufficient to keep only the lower powers of k. They assumed that this Taylor's series expansion existed,

$$\hat{c}(k) \cong \hat{c}(0) + \frac{d\hat{c}(0)}{dk} \, k + \frac{1}{2} \frac{d^2\hat{c}(0)}{dk^2} \, k^2$$

$$= c_0 - \frac{1}{6} \, c_2 k^2 \tag{5.46}$$

where

$$c_n \equiv \int d^3r\ r^n c(r) \tag{5.47}$$

From Eqs. (5.43) and (5.46)

$$[1 + \rho\hat{g}(k)]^{-1} = 1 - \rho\hat{c}(k) \cong 1 - \rho c_0 + \frac{1}{6}\rho c_2 k^2$$

$$= r_1^2[k^2 + \kappa_1^2] \tag{5.48}$$

where

$$\kappa_1^2 = \frac{1}{r_1^2}(1 - \rho c_0) \tag{5.49}$$

and

$$r_1^2 = \frac{1}{6}\rho c_2 \tag{5.50}$$

Therefore,

$$\hat{g}(k) = -\frac{1}{\rho} + \frac{1}{\rho r_1^2}\left[\frac{1}{k^2 + \kappa_1^2}\right] \tag{5.51}$$

and from Eq. (5.44)

$$g(r) = \frac{1}{4\pi\rho r_1^2} \frac{e^{-\kappa_1 r}}{r} \qquad \text{(large r)} \qquad (5.52)$$

Equation (5.52) is the classical OZ correlation function. Klein and Tisza have shown that as $T \to T_c$, $\kappa_1 \to 0$; thus when $T = T_c$, $g(r) \sim r^{-1}$ and the integral in Eq. (5.24) diverges as required. Experimentally it has been determined that κ_1, the inverse correlation length, is very temperature sensitive, while r_1 is a microscopic, relatively temperature insensitive length.

The single scattering inelastic DCS, Eq. (5.26), for critical nuclear scattering of thermal-neutrons from liquids now becomes, using the OZ correlation function, Eq. (5.52),

$$\left(\frac{d\sigma}{d\Omega}\right)_i^{(1)} = \frac{Na^2}{r_1^2}\left[\frac{1}{k^2 + \kappa_1^2}\right] \qquad \text{(small k)} \qquad (5.53)$$

i.e., a Lorentzian function of the wave vector transfer, $\vec{k}$. Since $k_i = k_f$ in the QA and $\vec{k} = \vec{k}_i - \vec{k}_f$,

$$k = 2k_i \sin\frac{\theta_S}{2} \qquad (5.54)$$

where θ_S is the scattering angle. For thermal-neutrons, $k_i \sim 1\ \text{Å}^{-1}$; thus, close to the critical point (κ_1 small), the cross section is very sharply peaked in the forward ($\theta_S = 0^\circ$) direction.

The small k restriction on the DCS, Eq. (5.53), which comes from the large r restriction on the OZ correlation function, Eq. (5.52),

is, in practice, unimportant for thermal-neutron scattering. For critical magnetic scattering, the spin correlations have the same OZ form as given by Eq. (5.52) and the single scattering magnetic DCS has the same form as Eq. (5.53), (see Chapter VII). In measuring the spin correlations in iron, Spooner and Averbach[15] have shown that the OZ correlation function is good for $r \gtrsim 15$ Å and that the single scattering magnetic DCS is good for $k^2 \lesssim 0.025$ Å^{-2}. Thus, from Eq. (5.54), for $k_i = 1$ Å^{-1}, the magnetic DCS is valid for scattering angles $\theta_S \lesssim 9^o$. For a correlation length of $\kappa_1^{-1} = 100$ Å, the magnetic DCS at $\theta_S = 9^o$ is already down to less than ½% of its peak ($\theta_S = 0^o$) value. Therefore, if sufficiently close to the critical point, the overwhelming portion of the scattering takes place within angles small enough to satisfy the small k restriction, thus effectively making Eq. (5.53) applicable without restriction.

In the next chapter, the contribution of double scattering to the critical nuclear cross section is treated.

CHAPTER VI

CRITICAL NUCLEAR DOUBLE SCATTERING FROM LIQUIDS

In this chapter, the double scattering cross section for critical nuclear scattering of thermal-neutrons from a liquid slightly above its critical temperature is obtained and compared to the single scattering cross section obtained in the last chapter.

In general, the double scattering DDCS for thermal-neutrons is given by Eq. (4.36),

$$\frac{d^2\sigma(2)}{d\Omega dE_f} = \lim_{\varepsilon\to 0} \frac{m_N^2}{(2\pi\hbar)^3} \frac{1}{\hbar^4} \frac{k_f}{k_i}$$

$$\times P_{12} \frac{d}{dt} \langle \vec{k}_{f_2} | \langle \vec{k}_{i_1} | \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t} dt_3 \int_{-\infty}^{t_3} dt_4$$

$$\times A_1(t_2)A_1(t_1)A_2(t_3)A_2(t_4) | \vec{k}_{i_2} \rangle | \vec{k}_{f_1} \rangle$$

$$\times \langle B(t_2)B(t_1)B(t_3)B(t_4) \rangle \, e^{\varepsilon(t_1 + t_2 + t_3 + t_4)} \qquad (6.1)$$

In applying this cross section to scattering from a liquid near its critical point, it may appear from the discussion in Chapter IV that the decomposition of the four B operator correlation function into products of pair correlation functions, as given in Eq. (4.37), is not

justified due to the long correlation lengths and relaxation times inherent in critical systems. However, this decomposition can still be made, albeit in a somewhat modified form.

For notational convenience, let

$$B(t_\ell) \equiv B_\ell \tag{6.2}$$

Now, consider the fluctuation of the operator B about its mean value, i.e.,

$$\tilde{B}_\ell = B_\ell - \langle B_\ell \rangle \tag{6.3}$$

It is easily shown that the four B operator correlation function in Eq. (6.1) can be written in terms of correlations of the fluctuations $\tilde{B}$ as

$$\langle B_2 B_1 B_3 B_4 \rangle = \langle \tilde{B}_2 \tilde{B}_1 \tilde{B}_3 \tilde{B}_4 \rangle + \mathcal{B}^{(3)} + \mathcal{B}^{(2)} \tag{6.4}$$

where

$$\mathcal{B}^{(3)} = \langle \tilde{B}_2 \tilde{B}_1 \tilde{B}_3 \rangle \langle B_4 \rangle + \langle \tilde{B}_2 \tilde{B}_1 \tilde{B}_4 \rangle \langle B_3 \rangle$$

$$+ \langle \tilde{B}_2 \tilde{B}_3 \tilde{B}_4 \rangle \langle B_1 \rangle + \langle \tilde{B}_1 \tilde{B}_3 \tilde{B}_4 \rangle \langle B_2 \rangle \tag{6.5}$$

and

$$\mathcal{B}^{(2)} = \langle \tilde{B}_2 \tilde{B}_1 \rangle \langle B_3 \rangle \langle B_4 \rangle + \langle \tilde{B}_2 \tilde{B}_3 \rangle \langle B_1 \rangle \langle B_4 \rangle$$

$$+ \langle \tilde{B}_2 \tilde{B}_4 \rangle \langle B_1 \rangle \langle B_3 \rangle + \langle \tilde{B}_1 \tilde{B}_3 \rangle \langle B_2 \rangle \langle B_4 \rangle$$

$$+ \langle \tilde{B}_1 \tilde{B}_4 \rangle \langle B_2 \rangle \langle B_3 \rangle + \langle \tilde{B}_3 \tilde{B}_4 \rangle \langle B_2 \rangle \langle B_1 \rangle$$

$$+ \langle B_2 \rangle \langle B_1 \rangle \langle B_3 \rangle \langle B_4 \rangle \tag{6.6}$$

Retaining only $\mathcal{B}^{(2)}$ in Eq. (6.4) would result in obtaining the purely elastic cross section; thus it will be dropped from further considerations. For nuclear scattering, as was shown previously, the operator B_ℓ is essentially the density operator at $\vec{r}_\ell$; thus $\tilde{B}$ is the density fluctuation operator. Near the critical point, the classical free energy for the liquid is a quadratic form in terms of density fluctuations, cf. Eq. (5.27). Therefore, as shown in Appendix II, a classical evaluation of the above correlations yields

$$\langle \tilde{B}_j \tilde{B}_k \tilde{B}_\ell \rangle = 0 \tag{6.7}$$

thus, $\mathcal{B}^{(3)} = 0$, and

$$\langle \tilde{B}_2 \tilde{B}_1 \tilde{B}_3 \tilde{B}_4 \rangle = \langle \tilde{B}_2 \tilde{B}_1 \rangle \langle \tilde{B}_3 \tilde{B}_4 \rangle + \langle \tilde{B}_2 \tilde{B}_3 \rangle \langle \tilde{B}_1 \tilde{B}_4 \rangle + \langle \tilde{B}_2 \tilde{B}_4 \rangle \langle \tilde{B}_1 \tilde{B}_4 \rangle \tag{6.8}$$

Therefore, for inelastic scattering only, Eq. (6.4) becomes

$$\langle B_2 B_1 B_3 B_4 \rangle = \langle \tilde{B}_2 \tilde{B}_1 \rangle \langle \tilde{B}_3 \tilde{B}_4 \rangle + \langle \tilde{B}_2 \tilde{B}_3 \rangle \langle \tilde{B}_1 \tilde{B}_4 \rangle + \langle \tilde{B}_2 \tilde{B}_4 \rangle \langle \tilde{B}_1 \tilde{B}_3 \rangle \tag{6.9}$$

which is the same decomposition as given by Eq. (4.37), except expressed in terms of pair correlations of the fluctuations of the operator B about its mean value. Thus, Eq. (4.46) is applicable for inelastic critical nuclear double scattering and yields for the inelastic DDCS

$$\left(\frac{d^2\sigma}{d\Omega dE_f}\right)_i^{(2)} = \lim_{\varepsilon \to 0} \frac{4m_N^2}{(2\pi\hbar)^8} \frac{1}{\hbar} \frac{k_f}{k_i}$$

$$\times g^4 \int d^3k_a \int d^3k_b \int d\omega'$$

$$\times \tilde{S}(\vec{k}_i - \vec{k}_a, \vec{k}_b - \vec{k}_i, \omega') \tilde{S}(\vec{k}_a - \vec{k}_f, \vec{k}_f - \vec{k}_b, \omega - \omega')$$

$$\times \left(k_a^2 - k_i^2 + \frac{2m_N\omega'}{\hbar} + i\varepsilon\right)^{-1} \left(k_b^2 - k_i^2 + \frac{2m_N\omega'}{\hbar} - i\varepsilon\right)^{-1} \tag{6.10}$$

where, from Eq. (5.2),

$$g = \frac{2\pi\hbar^2 a}{m_N} \tag{6.11}$$

and where the dynamic structure factor, $\tilde{S}$, involves correlations of density fluctuations

$$\tilde{S}(\vec{k}_2, \vec{k}_4, \omega') = \frac{1}{2\pi} \int d\tau \, e^{-i\omega'\tau} \langle [n_{\vec{k}_2}(0) - \langle n_{\vec{k}_2}(0) \rangle][n_{\vec{k}_4}(\tau) - \langle n_{\vec{k}_4}(\tau) \rangle] \rangle . \tag{6.12}$$

In the QA, the dynamic structure factor becomes

$$\tilde{S}(k_2,k_4,\omega') = \left[\langle n_{\vec{k}_2} n_{\vec{k}_4}\rangle - \langle n_{\vec{k}_2}\rangle\langle n_{\vec{k}_4}\rangle\right]\delta(\omega')$$

$$= \int d^3r_1 \int d^3r_2\, e^{-i\vec{k}_2\cdot\vec{r}_1}\, e^{-i\vec{k}_4\cdot\vec{r}_2}\, \Gamma(\vec{r}_1,\vec{r}_2)\delta(\omega') \qquad (6.13)$$

where the density-density correlation function, Γ, is given by Eq. (5.16).

At this point, the assumption that the liquid is translationally invariant will be made. However, some care must be exercised in making this assumption. Physically, translational invariance means that the liquid has the same properties at every point in the liquid, including points on or near the surface, i.e., bulk effects are homogeneous and boundary effects are neglected. Mathematically, the only time boundary effects are absent is when the target has an infinite spatial extent. Therefore, the assumption of translational invariance requires an infinite target if the mathematics is to remain <u>exactly</u> correct. However, unlike single scattering, double scattering, as will be shown, depends on target geometry; therefore, assuming an infinite target will lead to unphysical results. The finite nature of the target must be retained.

Proceeding formally, notice that the density-density correlation function, $\Gamma(\vec{r}_1,\vec{r}_2)$, is non-zero only when $\vec{r}_1$ and $\vec{r}_2$ are within the target. Therefore, the limits of the $\vec{r}_1$ and $\vec{r}_2$ integrations in Eq. (6.13) are confined to the target,

$$\tilde{S}(k_2,k_4,\omega') = \int_V d^3r_1 \int_V d^3r_2\, e^{-i\vec{k}_2\cdot\vec{r}_1}\, e^{-i\vec{k}_4\cdot\vec{r}_2}\, \Gamma(\vec{r}_1,\vec{r}_2)\delta(\omega') \qquad (6.14)$$

Now, assuming that the bulk effects of the target are homogeneous, Γ will depend only on the relative separation, $\vec{r} = \vec{r}_2 - \vec{r}_1$. Thus, making a change of integration variable, i.e., letting $\vec{r}_2 = \vec{r} + \vec{r}_1$,

$$\tilde{S}(\vec{k}_2, \vec{k}_4, \omega') =$$

$$\int_V d^3 r_1 \, e^{-i(\vec{k}_2 + \vec{k}_4)\cdot\vec{r}_1} \int_{\langle \vec{r}_1 \rangle} d^3 r \, e^{-i\vec{k}_4 \cdot \vec{r}} \, \Gamma(\vec{r}) \delta(\omega') \tag{6.15}$$

or, letting $\vec{r}_1 = \vec{r} - \vec{r}_2$, then replacing $\vec{r}_2$ with $\vec{r}_1$,

$$\tilde{S}(\vec{k}_2, \vec{k}_4, \omega') =$$

$$\int_V d^3 r_1 \, e^{-i(\vec{k}_2 + \vec{k}_4)\cdot\vec{r}_1} \int_{\langle \vec{r}_1 \rangle} d^3 r \, e^{i\vec{k}_2 \cdot \vec{r}} \, \Gamma(\vec{r}) \delta(\omega') \tag{6.16}$$

where, in both cases, the integral over $\vec{r}$ depends on $\vec{r}_1$, and $\Gamma(\vec{r})$ is given by Eq. (5.23). Now, when the liquid is slightly above its critical temperature (on the critical isochore), $\Gamma(\vec{r})$ will have a microscopic range, i.e., $\kappa_1^{-1} \sim 10\ \text{Å} - 10^3\ \text{Å}$. Therefore, the only place this dependence on $\vec{r}_1$ is important is within a microscopic distance of the surface. Neglecting this boundary effect, $\tilde{S}$ can be written, from Eq. (6.15),

$$\tilde{S}(\vec{k}_2, \vec{k}_4, \omega') = \int_V d^3 r_1 \, e^{-i(\vec{k}_2 + \vec{k}_4)\cdot\vec{r}_1} \int_{-\infty}^{\infty} d^3 r \, e^{-i\vec{k}_4 \cdot \vec{r}} \, \Gamma(\vec{r}) \delta(\omega') \tag{6.17}$$

or, from Eq. (6.16),

$$\tilde{S}(\vec{k}_2,\vec{k}_4,\omega') = \int d^3r_1\, e^{-i(\vec{k}_2+\vec{k}_4)\cdot\vec{r}_1} \int_{-\infty}^{\infty} d^3r\, e^{i\vec{k}_2\cdot\vec{r}}\, \Gamma(\vec{r})\,\delta(\omega') \qquad (6.18)$$

Mathematically, the equality sign holds in Eqs. (6.17) and (6.18) and these two expressions for $\tilde{S}$ are equivalent only for an infinite target, since then

$$\int_{-\infty}^{\infty} d^3r_1\, e^{-i(\vec{k}_2+\vec{k}_4)\cdot\vec{r}_1} = (2\pi)^3\delta(\vec{k}_2+\vec{k}_4) \qquad (6.19)$$

thus, $\vec{k}_2 = -\vec{k}_4$ in order for $\vec{S}$ to be non-zero. However, for a macroscopic target, these two expressions for $\vec{S}$ are still physically equivalent, since the integral over $\vec{r}_1$ will require

$$|\vec{k}_2+\vec{k}_4| \lesssim \ell^{-1} \qquad (6.20)$$

where ℓ is a linear dimension of the target. Thus, for thermal-neutrons $\vec{k}_2 \cong -\vec{k}_4$.

Using Eq. (5.23) for $\Gamma(r)$ and dropping the self-correlation term,[16] from Eqs. (6.11), (6.17) and (6.18) the double scattering inelastic DDCS, Eq. (6.10), becomes

$$\left(\frac{d\sigma}{d\Omega dE_f}\right)_i^{(2)} = \lim_{\varepsilon\to 0} \frac{\rho^4 a^4}{4\pi^4} \frac{1}{\hbar}$$

$$\times \int d^3k_a \int d^3k_b \int_V d^3r_1 \int_V d^3r_2 \; e^{-i(\vec{k}_a-\vec{k}_b)\cdot(\vec{r}_2-\vec{r}_1)}$$

$$\times \int d^3r \int d^3r' \; e^{i(\vec{k}_a-\vec{k}_f)\cdot\vec{r}} \; e^{-i(\vec{k}_b-\vec{k}_i)\cdot\vec{r}'} \; g(r)g(r')$$

$$\times (k_a^2-k_i^2+i\varepsilon)^{-1} (k_b^2-k_i^2-i\varepsilon)^{-1}\delta(\omega) \tag{6.21}$$

where Eqs. (5.6) and (5.8) have been utilized.

The $\vec{k}_a$ and $\vec{k}_b$ integrations in Eq. (6.21) are easily evaluated using the methods of complex contour integration and yield

$$\lim_{\varepsilon\to 0} \int d^3k_a \int d^3k_b \; e^{-i\vec{k}_a\cdot(\vec{r}_2-\vec{r}_1-\vec{r})} \; e^{i\vec{k}_b\cdot(\vec{r}_2-\vec{r}_1-\vec{r}')}$$

$$\times (k_a^2-k_i^2+i\varepsilon)^{-1}(k_b^2-k_i^2-i\varepsilon)^{-1}$$

$$= 4\pi^4 \frac{e^{-ik_i|\vec{r}_2-\vec{r}_1-\vec{r}|}}{|\vec{r}_2-\vec{r}_1-\vec{r}|} \frac{e^{ik_i|\vec{r}_2-\vec{r}_1-\vec{r}'|}}{|\vec{r}_2-\vec{r}_1-\vec{r}'|} \tag{6.22}$$

From Eq. (6.21), the double scattering inelastic DCS becomes

$$\left(\frac{d\sigma}{d\Omega}\right)_i^{(2)} = \rho^4 a^4 \int_V d^3r_1 \int_V d^3r_2 \int d^3r \int d^3r'$$

$$\times e^{i\vec{k}_i\cdot\vec{r}'} e^{-i\vec{k}_f\cdot\vec{r}} g(r)g(r')$$

$$\times \frac{e^{-ik_i|\vec{r}_2-\vec{r}_1-\vec{r}|}}{|\vec{r}_2-\vec{r}_1-\vec{r}|} \frac{e^{ik_i|\vec{r}_2-\vec{r}_1-\vec{r}'|}}{|\vec{r}_2-\vec{r}_1-\vec{r}'|} \tag{6.23}$$

Since $\vec{r}$ and $\vec{r}'$ appear in the correlation function, g, they are restricted to microscopic distances by the correlation range, κ_1^{-1}, i.e., $r \lesssim \kappa_1^{-1}$. Therefore, since $\vec{r}_1$ and $\vec{r}_2$ are free to range over the entire target which is of macroscopic dimensions, the overwhelming contributions to the integrals over $\vec{r}_1$ and $\vec{r}_2$ come when

$$|\vec{r}_2-\vec{r}_1| \ggg r, r'$$

Thus,

$$|\vec{r}_2-\vec{r}_1-\vec{r}| \simeq r_{21} - \hat{r}_{21}\cdot\vec{r} \tag{6.24}$$

$$|\vec{r}_2-\vec{r}_1-\vec{r}'| \simeq r_{21} - \hat{r}_{21}\cdot\vec{r}' \tag{6.25}$$

where $\vec{r}_{21} = \vec{r}_2 - \vec{r}_1$ and $\hat{r}_{21} = \vec{r}_{21}/r_{21}$. The exponential terms in Eq. (6.23) become

$$\frac{e^{-ik_i|\vec{r}_2-\vec{r}_1-\vec{r}|}}{|\vec{r}_2-\vec{r}_1-\vec{r}|} \simeq \frac{e^{-ik_i r_{21}}\, e^{i\vec{q}\cdot\vec{r}}}{r_{21}} \tag{6.26}$$

$$\frac{e^{ik_i|r_2-r_1-r'|}}{|r_2-r_1-r'|} \simeq \frac{e^{ik_i r_{21}}\, e^{-i\vec{q}\cdot\vec{r}'}}{r_{21}} \tag{6.27}$$

where

$$\vec{q} = k_i \hat{r}_{21} \tag{6.28}$$

represents the wave vector of magnitude k_i along the direction $\hat{r}_{21}$. Identifying $\vec{r}_1$ as locating the first scattering event and $\vec{r}_2$ as locating the second scattering event, $\vec{q}$ is the intermediate wave vector of the neutron between scattering events.

From Eqs. (6.26) and (6.27), the double scattering inelastic DCS, Eq. (6.23), becomes, after a change of integration variable,

$$\left(\frac{d\sigma}{d\Omega}\right)_i^{(2)} = \rho^4 a^4 \int_V d^3r_1 \int_{\langle\vec{r}_1\rangle} d^3r_{21}\, r_{21}^{-2}\, \hat{g}(\vec{k}_i-\vec{q})\hat{g}(\vec{q}-\vec{k}_f) \tag{6.29}$$

where

$$\hat{g}(\vec{k}_1) = \int d^3r\, e^{i\vec{k}_1\cdot\vec{r}}\, g(r) \tag{6.30}$$

is the Fourier transform of g(r). The limits of integration on $\vec{r}_{21}$

still go over the target but now depend on $\vec{r}_1$.

Using the OZ correlation function for g(r), Eq. (5.52), the double scattering critical nuclear inelastic DCS, Eq. (6.29), becomes

$$\left(\frac{d\sigma}{d\Omega}\right)_i^{(2)} = C \int_V d^3r_1 \int_{\langle \vec{r}_1 \rangle} d^3r_{21}\, r_{21}^{-2}\left[|\vec{k}_i-\vec{q}|^2 + \kappa_1^2\right]^{-1}\left[|\vec{q}-\vec{k}_f|^2 + \kappa_1^2\right]^{-1} \quad (6.31)$$

where

$$C = \frac{\rho^2 a^4}{r_1^4} \quad (6.32)$$

[the r_1 contained in C is the OZ "direct correlation length" defined by Eq. (5.50)]. Remembering that $\vec{q}$ depends on $\hat{r}_{21}$, it becomes evident that the double scattering depends on target geometry. Now, using Eq. (6.28) for $\vec{q}$, the magnitudes of the intermediate wave vector transfers are

$$|\vec{k}_i-\vec{q}| = 2k_i \sin\frac{\theta_1}{2} \quad (6.33)$$

$$|\vec{q}-\vec{k}_f| = 2k_i \sin\frac{\theta_2}{2} \quad (6.34)$$

where the intermediate scattering angles θ_1 and θ_2 are depicted in Figure 1. As the critical temperature is approached, $\kappa_1 \to 0$, the DCS, Eq. (6.31), is very sharply peaked about $\theta_1 = \theta_2 = 0$ and $\hat{r}_{21}$ lies in a

small solid angle centered about $\vec{k}_i$ with vertex at $\vec{r}_1$. Therefore, defining $\ell(\vec{r}_1)$ as the length from $\vec{r}_1$ in the direction of $\vec{k}_i$ to the boundary of the target (see Figure 1), to a very good approximation, the DCS, Eq. (6.31), can be written

$$\left(\frac{d\sigma}{d\Omega}\right)_i^{(2)} = C \int_V d^3r_1 \, \ell(\vec{r}_1) \int_0^{2\pi} d\phi_1 \int_0^{\pi} d\theta_1 \sin\theta_1$$

$$\times \left[4k_i^2 \sin^2\frac{\theta_1}{2} + \kappa_1^2\right]^{-1} \left[4k_i^2 \sin^2\frac{\theta_2}{2} + \kappa_1^2\right]^{-1}$$

$$\equiv C \int_V d^3r_1 \, \ell(\vec{r}_1) G(k_i, \theta_S, \kappa_1) \tag{6.35}$$

The angle θ_2 will depend on ϕ_1, θ_1 and the scattering angle θ_S. Defining

$$\langle \ell \rangle \equiv V^{-1} \int_V d^3r_1 \, \ell(\vec{r}_1) \tag{6.36}$$

the double scattering critical nuclear inelastic DCS, Eq. (6.35), from Eq. (6.32), becomes

$$\left(\frac{d\sigma}{d\Omega}\right)_i^{(2)} = N \frac{\rho a^4}{r_1^4} \langle \ell \rangle \, G(k_i, \theta_S, \kappa_1) \tag{6.37}$$

The length $\langle \ell \rangle$ will be of the order of magnitude of the linear dimension of the target. For a spherical target of radius R, $\langle \ell \rangle = 3R/4$ and for a slab of thickness L, $\langle \ell \rangle = L/2$.

The integral $G(k_i,\theta_S,\kappa_1)$, defined by Eq. (6.35), is evaluated in Appendix III in the small angle approximation, i.e., $\sin\theta \cong \theta$ and is given by

$$G = \frac{\pi}{k_i^2\kappa_1^2 x(x^2+4)^{\frac{1}{2}}} \ln\left[\frac{x^3+3x+(x^2+1)(x^2+4)^{\frac{1}{2}}}{(x^2+4)^{\frac{1}{2}}-x}\right] \tag{6.38}$$

where

$$x = k\kappa_1^{-1} \tag{6.39}$$

and

$$k = k_i\theta_S \tag{6.40}$$

is the wave vector transfer of the neutron. In the same small angle approximation, the single scattering critical nuclear inelastic DCS, Eq. (5.53), is

$$\left(\frac{d\sigma}{d\Omega}\right)_i^{(1)} = N\frac{a^2}{r_1^2\kappa_1^2}(1+x^2)^{-1} \tag{6.41}$$

Therefore, the combined inelastic DCS, single plus double scattering, from Eqs. (6.37) and (6.41), is given by

$$\left(\frac{d\sigma}{d\Omega}\right)_i = \left(\frac{d\sigma}{d\Omega}\right)_i^{(1)} + \left(\frac{d\sigma}{d\Omega}\right)_i^{(2)} = \left(\frac{d\sigma}{d\Omega}\right)_i^{(1)} [1 + \beta H(x)] \tag{6.42}$$

where

$$\beta = \frac{\pi\rho\langle\ell\rangle a^2}{k_i^2 r_i^2} \tag{6.43}$$

and

$$H(x) = \frac{x^2+1}{x(x^2+4)^{\frac{1}{2}}} \ln\left[\frac{x^3+3x+(x^2+1)(x^2+4)^{\frac{1}{2}}}{(x^2+4)^{\frac{1}{2}}-x}\right] \tag{6.44}$$

Since $H(0) = 1$, β is the fraction of double scattering at $x = 0$. Notice that the double scattering will become more pronounced, i.e., β will increase as (1) ρ increases, (2) the target size increases ($\langle\ell\rangle$ increases), and (3) the neutron's energy, E_i, decreases (k_i^2 decreases). In fact, double scattering will be more sensitive to the neutron's initial wave vector (β going like k_i^{-2}) than to target size.

Returning to the decomposition of the four B operator correlation function given in Eq. (6.9), the first two terms, which, up to now, have been neglected, are now considered. The second term in Eq. (6.9), $\langle\tilde{B}_2\tilde{B}_3\rangle\langle\tilde{B}_1\tilde{B}_4\rangle$, gives rise to a DCS for critical nuclear double scattering that represents a quantum interference effect and can be neglected for macroscopic targets. Explicitly, it gives for the DCS,

$$\left(\frac{d\sigma}{d\Omega}\right)_i^{(2)} = \rho^4 a^4 \int_V d^3r_1 \int_{\langle\vec{r}_1\rangle} d^3r_{21}\, r_{21}^{-2}\, e^{i(\vec{k}_i+\vec{k}_f)\cdot\vec{r}_{21}}\, \hat{g}(\vec{k}_i-\vec{q})\hat{g}(\vec{q}-\vec{k}_f) \tag{6.45}$$

which is identical to the DCS given by Eq. (6.29) except for the phase factor

$$e^{i(\vec{k}_i+\vec{k}_f)\cdot\vec{r}_{21}}$$

In terms of order of magnitude, Eq. (6.45) goes like $(k_i \ell)^{-2} \left(\frac{d\sigma}{d\Omega}\right)_i^{(2)}$, where ℓ represents a linear dimension of the target and $\left(\frac{d\sigma}{d\Omega}\right)_i^{(2)}$ is the DCS given by Eq. (6.37).

The first term in Eq. (6.9), $\langle \tilde{B}_2 \tilde{B}_1 \rangle \langle \tilde{B}_3 \tilde{B}_4 \rangle$, gives rise to a DCS for critical nuclear double scattering that represents an elastic diffraction effect and is essentially all forward scattering for macroscopic targets. Explicitly, it gives for the DCS

$$\left(\frac{d\sigma}{d\Omega}\right)^{(2)} = \left| \rho^2 a^2 \int d^3 r_1 \, e^{-i\vec{k}\cdot\vec{r}_1} \int d^3 r \, g(r) \, e^{i\vec{k}_f\cdot\vec{r}} \, \frac{e^{-ik_i r}}{r} \right|^2 \qquad (6.46)$$

This cross section is non-negligible only for scattering angles $\theta_S \lesssim (k_i \ell)^{-1}$.

Numerical results for double scattering are considered in detail only for critical magnetic scattering from ferromagnets (see Chapter IX), since much more experimental effort has been invested in this area than has been invested in critical nuclear scattering of neutrons from liquids. However, for currently employed ranges of experimental parameters, it does appear that double scattering is a small effect in critical nuclear scattering from liquids. In a recent experiment on

critical scattering from neon,[17] the double scattering parameter is estimated to be $\beta \triangleq 0.002$.

In the next chapter, critical magnetic scattering from ferromagnets slightly above their Curie temperature is considered.

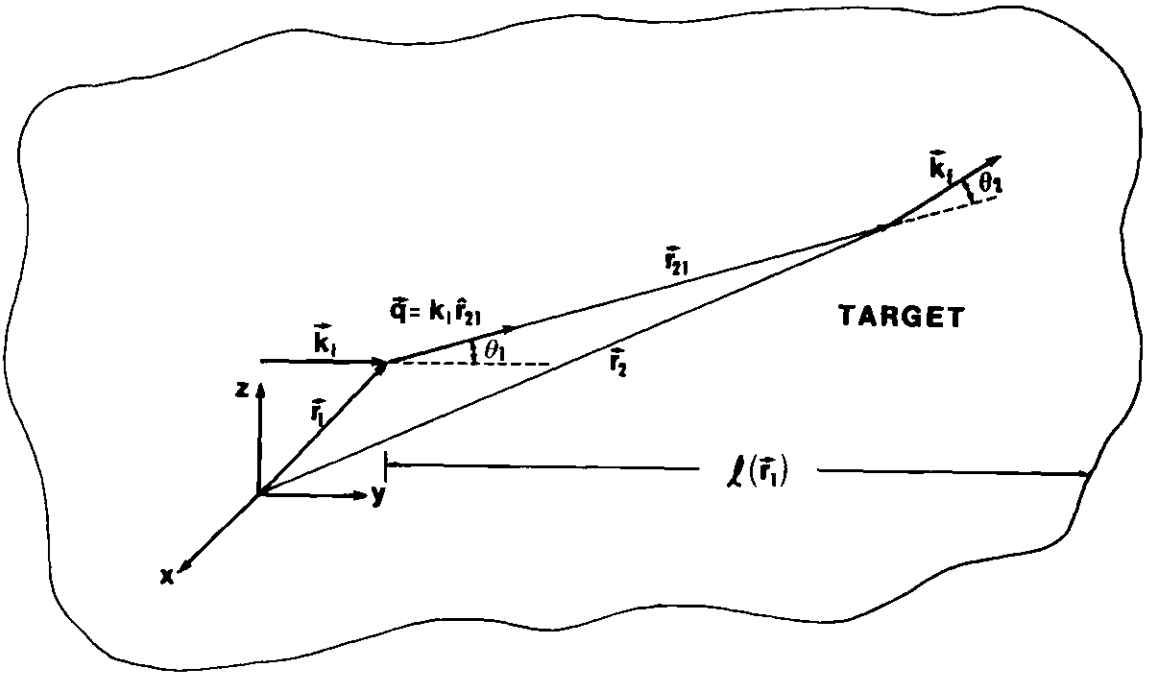

Figure 1. Double Scattering Geometry.

CHAPTER VII

CRITICAL MAGNETIC SINGLE SCATTERING FROM FERROMAGNETS

In this chapter, the single scattering cross section for critical magnetic scattering of thermal-neutrons from a ferromagnet slightly above its Curie temperature, T_c, is obtained.

The Magnetic Interaction

Letting the target be a ferromagnetic crystal, only the case of magnetic scattering, where the spin of the incoming neutron couples to the spin of the unpaired target electrons, is considered. In addition, only ferromagnets for which the orbital contribution to the electron's magnetic (dipole) moment is negligible or can simply be taken account of by adjusting the electronic spin quantum number, are treated.

The interaction between the neutron and the electrons of the target is given by

$$\phi(t) = \sum_j \phi[\vec{r}(t), \vec{\mu}_N, \vec{\mu}_j(t)] \tag{7.1}$$

where the interaction, $\phi(\vec{r}, \vec{\mu}_N, \vec{\mu}_j)$, between the magnetic moment, $\vec{\mu}_N$, of the neutron at $\vec{r}_N$ and the magnetic moment, $\vec{\mu}_j$, of the j^{th} electron at $\vec{r}_j$, is given by

$$\phi(\vec{r}, \vec{\mu}_N, \vec{\mu}_j) = - \vec{\mu}_j \cdot \vec{H}_N(\vec{r}, \vec{\mu}_N) \tag{7.2}$$

Here, $\vec{r} = \vec{r}_N - \vec{r}_j$ and $\vec{H}_N$ is the magnetic field due to the magnetic moment of the neutron. By definition of the vector potential, $\vec{A}_N$,

$$\vec{H}_N \equiv \vec{\nabla} \times \vec{A}_N \tag{7.3}$$

and $\vec{A}_N$ due to a magnetic moment $\vec{\mu}_N$ is well-known to be[18]

$$\vec{A}_N = \vec{\mu}_N \times \frac{\vec{r}}{r^3} \tag{7.4}$$

Thus, the magnetic field, Eq. (7.3), becomes

$$\vec{H}_N = \vec{\nabla} \times \left[\vec{\mu}_N \times \frac{\vec{r}}{r^3}\right] = - \vec{\nabla} \times \left[\vec{\mu}_N \times \vec{\nabla}\left(\frac{1}{r}\right)\right] \tag{7.5}$$

where the identity[19]

$$\vec{\nabla}\left(\frac{1}{r}\right) = - \frac{\vec{r}}{r^3} \tag{7.6}$$

has been used. Using the vector relation[19]

$$\vec{\nabla} \times [\vec{A} \times \vec{B}] = \vec{A}(\vec{\nabla} \cdot \vec{B}) - \vec{B}(\vec{\nabla} \cdot \vec{A}) + (\vec{B} \cdot \vec{\nabla})\vec{A} - (\vec{A} \cdot \vec{\nabla})\vec{B} \tag{7.7}$$

Eq. (7.5) becomes

$$\vec{H}_N = (\vec{\mu}_N \cdot \vec{\nabla})\vec{\nabla}\left(\frac{1}{r}\right) + 4\pi\vec{\mu}_N\delta(\vec{r}) \tag{7.8}$$

where the identity[20]

$$\nabla^2\left(\frac{1}{r}\right) = -\,4\pi\delta(\vec{r}) \tag{7.9}$$

has been used. Now, using the vector relation[19]

$$\vec{\nabla}(\vec{A}\cdot\vec{B}) = \vec{A}\times(\vec{\nabla}\times\vec{B}) + \vec{B}\times(\vec{\nabla}\times\vec{A}) + (\vec{B}\cdot\vec{\nabla})\vec{A} + (\vec{A}\cdot\vec{\nabla})\vec{B}$$

one obtains

$$\vec{\nabla}\left[\vec{\mu}_N\cdot\vec{\nabla}\left(\frac{1}{r}\right)\right] = (\vec{\mu}_N\cdot\vec{\nabla})\vec{\nabla}\left(\frac{1}{r}\right) \tag{7.11}$$

since

$$\vec{\nabla}\times\vec{\nabla}\left(\frac{1}{r}\right) \equiv 0 \tag{7.12}$$

Therefore, the interaction, Eq. (7.2), from Eqs. (7.8) and (7.11), can be written

$$\phi(\vec{r},\vec{\mu}_N,\vec{\mu}_j) = -\,\vec{\mu}_j\cdot\vec{\nabla}\left[\vec{\mu}_N\cdot\vec{\nabla}\left(\frac{1}{r}\right)\right] - 4\pi\vec{\mu}_j\cdot\vec{\mu}_N\delta(\vec{r}) \tag{7.13}$$

The Fourier transform of ϕ, $\hat{\phi}$, Eq. (4.28), is given by

$$\hat{\phi}(\vec{k}_1,\vec{\mu}_N,\vec{\mu}_j) = \int d^3r\, e^{-i\vec{k}_1\cdot\vec{r}} \left\{-\vec{\mu}_j \cdot \vec{\nabla}\left[\vec{\mu}_N \cdot \vec{\nabla}\left(\frac{1}{r}\right)\right] - 4\pi\vec{\mu}_j \cdot \vec{\mu}_N\delta(\vec{r})\right\}$$

$$= -\int d^3r\, e^{-i\vec{k}_1\cdot\vec{r}}\, \vec{\mu} \cdot \vec{\nabla}\left[\vec{\mu}_N \cdot \vec{\nabla}\left(\frac{1}{r}\right)\right] - 4\pi\vec{\mu}_j \cdot \vec{\mu}_N \qquad (7.14)$$

As shown in Appendix IV,

$$-\int d^3r\, e^{-i\vec{k}_1\cdot\vec{r}}\, \vec{\mu}_j \cdot \vec{\nabla}\left[\vec{\mu}_N \cdot \vec{\nabla}\left(\frac{1}{r}\right)\right] = 4\pi(\vec{\mu}_N \cdot \hat{k}_1)(\vec{\mu}_j \cdot \hat{k}_1) \qquad (7.15)$$

where $\hat{k}_1 = \vec{k}_1/k_1$. Therefore,

$$\hat{\phi}(\vec{k}_1,\vec{\mu}_N,\vec{\mu}_j) = -4\pi\vec{\mu}_N \cdot\left[\vec{\mu}_j - \hat{k}_1(\vec{\mu}_j \cdot \hat{k}_1)\right] = \vec{a}(\vec{\mu}_N) \cdot \vec{b}(\vec{k}_1,\vec{\mu}_j) \qquad (7.16)$$

where

$$\vec{a}(\mu_N) = -4\pi\vec{\mu}_N \qquad (7.17)$$

$$\vec{b}(\vec{k}_1,\vec{\mu}_j) = \vec{\mu}_j - \hat{k}_1(\vec{\mu}_j \cdot \hat{k}_1) \qquad (7.18)$$

are the operators first introduced in Eq. (4.29). From Eq. (7.16), the interaction $\phi(t)$, Eq. (4.27), can now be written in the desired form, Eq. (4.2),

$$\phi(t) = \sum_{\vec{k}_1} A^{\alpha}(\vec{k}_1,t)B^{\alpha}(\vec{k}_1,t) \qquad (7.19)$$

where

$$A^{\alpha}(\vec{k}_1,t) = e^{i\vec{k}_1\cdot\vec{r}_N(t)}\, a^{\alpha}(\vec{\mu}_N) \tag{7.20}$$

$$B^{\alpha}(\vec{k}_1,t) = \sum_j e^{-i\vec{k}_1\cdot\vec{r}_j(t)}\, b^{\alpha}\left[\vec{k}_1,\vec{\mu}_j(t)\right] \tag{7.21}$$

The magnetic moments of the neutron and the electron are related to their respective spins, $\vec{s}_N$ and $\vec{s}_j$, by

$$\vec{\mu}_N = g_N\mu_0\vec{s}_N \tag{7.22}$$

$$\vec{\mu}_j = -g_e\mu_B\vec{s}_j \tag{7.23}$$

where

$$\mu_0 = \frac{e\hbar}{2m_N c} \tag{7.24}$$

is the nuclear magneton,

$$\mu_B = \frac{e\hbar}{2m_e c} \tag{7.25}$$

is the Bohr magneton, g_N and g_e are, respectively, the neutron's and electron's Landé g-factor, m_e is the electron's mass and e is the magnitude of the electron's charge. Experimentally,

$$g_N \cong -3.82 \tag{7.26}$$

$$g_e \cong 2.00 \tag{7.27}$$

Therefore,

$$\vec{\mu}_N = -g\frac{e\hbar}{m_N c}\vec{s}_N \tag{7.28}$$

$$\vec{\mu}_j = -\frac{e\hbar}{m_e c}\vec{s}_j \tag{7.29}$$

where $g = -g_N/2 \cong 1.91$. Notice, that since the z-component of the neutron's spin is $\frac{1}{2}$, i.e., $s_N^z = \frac{1}{2}$, the z-component of the neutron's magnetic moment is

$$\mu_N^z = g\frac{e\hbar}{2m_N c} = g\mu_0 \tag{7.30}$$

thus, g is the z-component of the neutron's magnetic moment in nuclear magnetons. Replacing these magnetic moments, Eqs. (7.28) and (7.29), in Eqs. (7.17) and (7.18) gives

$$\vec{a}(\vec{s}_N) = 4\pi g\frac{e\hbar}{m_N c}\vec{s}_N \tag{7.31}$$

$$\vec{b}\left[\vec{k}_1, \vec{s}_j(t)\right] = -\frac{e\hbar}{m_e c}\left\{\vec{s}_j(t) - \hat{k}_1\left[\vec{s}_j(t)\cdot\hat{k}_1\right]\right\} \tag{7.32}$$

The Magnetic Dynamic Structure Factor

The single scattering magnetic DDCS is given by Eq. (4.33),

$$\frac{d^2\sigma(1)}{d\Omega dE_f} = \frac{m_N^2}{4\pi^2\hbar^5}\frac{k_f}{k_i} M_N^{\alpha\beta} S^{\alpha\beta}(\vec{k},\omega) \tag{7.33}$$

where the neutron matrix elements, $M_N^{\alpha\beta}$, are given by Eq. (4.34), and the dynamic structure factor, $S^{\alpha\beta}(\vec{k},\omega)$, is given by Eq. (4.35), which, from Eqs. (7.21) and (7.32), now becomes

$$S^{\alpha\beta}(\vec{k},\omega) =$$

$$\frac{1}{2\pi}\left(\frac{e\hbar}{m_e c}\right)^2 \int d\tau\, e^{-i\omega\tau} \left\langle \sum_j e^{-i\vec{k}\cdot\vec{r}_j(0)} p_j^{\alpha}(\vec{k},0) \sum_\ell e^{i\vec{k}\cdot\vec{r}_\ell(\tau)} p_\ell^{\beta}(\vec{k},\tau) \right\rangle \tag{7.34}$$

where

$$\vec{p}_j = \vec{s}_j - \hat{k}(\vec{s}_j \cdot \hat{k}) \tag{7.35}$$

is the component of $\vec{s}_j$ perpendicular to $\vec{k}$. Again, the sums over j and ℓ in the target average in Eq. (7.34) are over the unpaired electrons of the target. Assuming the target to be made up of identical atoms, with n unpaired electrons per atom and one atom per unit cell, the target average in Eq. (7.34) is

$$\langle \cdots \rangle = \Big\langle \sum_{j,\ell=1}^{N} e^{-i\vec{k}\cdot\vec{R}_j(0)} \, e^{i\vec{k}\cdot\vec{R}_\ell(\tau)} \sum_{j',\ell'=1}^{n}$$

$$\times \, e^{-i\vec{k}\cdot\vec{u}_{jj'}(0)} \, e^{i\vec{k}\cdot\vec{u}_{\ell\ell'}(\tau)} \, p^{\alpha}_{jj'}(\vec{k},0) p^{\beta}_{\ell\ell'}(\vec{k},\tau) \Big\rangle \qquad (7.36)$$

where $\vec{R}_j$ is the position vector of the nucleus of the j^{th} atom, $\vec{u}_{jj'}$ is the position vector of the j'^{th} electron of the j^{th} atom, relative to $\vec{R}_j$, and N is the number of atoms in the target. Neglecting nuclear thermal motions, i.e., assuming a rigid lattice, $\vec{R}_j(\tau) \cong \vec{R}_j(0)$, a fixed c-number. The target average can now be written as

$$\langle \cdots \rangle = \sum_{j,\ell} e^{-i\vec{k}\cdot\vec{R}_j} \, e^{i\vec{k}\cdot\vec{R}_\ell}$$

$$\times \Big\langle \sum_{j'\ell'} e^{-i\vec{k}\cdot\vec{u}_{jj'}(0)} \, e^{i\vec{k}\cdot\vec{u}_{\ell\ell'}(\tau)} \, p^{\alpha}_{jj'}(\vec{k},0) p^{\beta}_{\ell\ell'}(\vec{k},\tau) \Big\rangle \qquad (7.37)$$

where the average is now over the target electronic spatial and spin states.

For most temperatures of interest,[21] the initial electronic spatial state will be the ground state, $|\Phi_o\rangle$. Therefore, neglecting any coupling between the electronic spatial and spin states

$$\langle \cdots \rangle = \sum_{j,\ell} e^{-i\vec{k}\cdot\vec{R}_j} \, e^{i\vec{k}\cdot\vec{R}_\ell} \sum_{\sigma_i^T} P_{\sigma_i^T} \qquad (7.38)$$

$$\times \, \langle \sigma_i^T | \langle \Phi_o | \sum_{j',\ell'} e^{-i\vec{k}\cdot\vec{u}_{jj'}(0)} \, e^{i\vec{k}\cdot\vec{u}_{\ell\ell'}(\tau)} \, p^{\alpha}_{jj'}(\vec{k},0) p^{\beta}_{\ell\ell'}(\vec{k},\tau) | \Phi_o \rangle | \sigma_i^T \rangle$$

where $P_{\sigma_i^T}$ is the probability of the occurrence of the target spin state $|\sigma_i^T\rangle$. Inserting a complete set of electronic spatial states in Eq. (7.38), all matrix elements that involve transitions to excited electronic spatial states are neglected, since thermal-neutrons have insufficient energies to cause such transitions.[22] Therefore,

$$\langle \cdots \rangle = \sum_{j,\ell} e^{-i\vec{k}\cdot\vec{R}_j} e^{i\vec{k}\cdot\vec{R}_\ell} \sum_{\sigma_i^T} P_{\sigma_i^T}$$

$$\times \langle \sigma_i^T | \langle \Phi_o | \sum_{j'} e^{-i\vec{k}\cdot\vec{u}_{jj'}(0)} p^{\alpha}_{jj'}(\vec{k},0) | \Phi_o \rangle$$

$$\times \langle \Phi_o | \sum_{\ell'} e^{i\vec{k}\cdot\vec{u}_{\ell\ell'}(0)} p^{\beta}_{\ell\ell'}(\vec{k},\tau) | \Phi_o \rangle | \sigma_i^T \rangle \qquad (7.39)$$

Assuming that the electronic spin states are adequately described by the Heisenberg model, which ascribes to each atom an effective spin operator, $\vec{S}$, of fixed length, implies that the electronic spatial wavefunctions are localized around each atom; thus overlapping between wavefunctions associated with different atoms can be neglected, i.e.,

$$|\Phi_o\rangle = |\phi_1\rangle|\phi_2\rangle \cdots |\phi_N\rangle \qquad (7.40)$$

where $\langle \vec{r} | \phi_\ell \rangle$ is the ground state electronic spatial wavefunction associated with the ℓ^{th} atom. Therefore, from Eq. (7.39),

$$\langle \Phi_o | \sum_{j'} e^{-i\vec{k}\cdot\vec{u}_{jj'}} p^{\alpha}_{jj'} | \Phi_o \rangle = \langle \phi_1 | \cdots \langle \phi_N | \sum_{j'} e^{-i\vec{k}\cdot\vec{u}_{jj'}} p^{\alpha}_{jj'} | \phi_N \rangle \cdots | \phi_1 \rangle$$

$$= \langle \phi_j | \sum_{j'} e^{-i\vec{k}\cdot\vec{u}_{jj'}} | \phi_j \rangle p^{\alpha}_{jj'} = \langle \phi | \sum_{j'} e^{-i\vec{k}\cdot\vec{u}_{j'}} | \phi \rangle p^{\alpha}_{jj'} \qquad (7.41)$$

The last equation in Eq. (7.41) follows from the fact that the electronic spatial matrix element is independent of the particular atom chosen, since all atoms are assumed to be identical. From Eq. (7.41),

$$\langle \phi | \sum_{j'} e^{-i\vec{k}\cdot\vec{u}_{j'}} | \phi \rangle p^{\alpha}_{jj'} = \int d^3u \, e^{-i\vec{k}\cdot\vec{u}} \langle \phi | \sum_{j'} \delta(\vec{u}-\vec{u}_{j'}) | \phi \rangle p^{\alpha}_{jj'} \qquad (7.42)$$

Now,

$$p^{\alpha}_{j1} = p^{\alpha}_{j2} = \cdots = p^{\alpha}_{jn} \equiv p^{\alpha}_{j} \qquad (7.43)$$

since all n unpaired spins are aligned in the same direction (Hund's rule). Thus, Eq. (7.42) becomes

$$\langle \phi | \sum_{j'} e^{-i\vec{k}\cdot\vec{u}_{j'}} | \phi \rangle p^{\alpha}_{jj'} = \int d^3u \, e^{-i\vec{k}\cdot\vec{u}} \delta'(\vec{u}) p^{\alpha}_{j} \qquad (7.44)$$

where

$$\delta'(\vec{u}) = \langle \phi | \sum_{j'} \delta(\vec{u}-\vec{u}_{j}) | \phi \rangle \qquad (7.45)$$

is the atomic, unpaired electron spin density. Multiplying and dividing Eq. (7.44) by n, one obtains

$$\langle\phi| \sum_{j'} e^{-i\vec{k}\cdot\vec{u}_{j'}} |\phi\rangle p^{\alpha}_{jj'} = \int d^3u\, e^{-i\vec{k}\cdot\vec{u}}\, \delta(\vec{u}) P^{\alpha}_{j} \tag{7.46}$$

where $\delta(\vec{u}) = \delta'(\vec{u})/n$ is the normalized spin density,

$$P^{\alpha}_{j} = np^{\alpha}_{j} = \left[\vec{S}_j - \hat{k}(\vec{S}_j \cdot \hat{k})\right]^{\alpha} \tag{7.47}$$

and

$$\vec{S}_j = n\vec{s}_j \tag{7.48}$$

is the effective Heisenberg spin operator of the j^{th} atom.[23] From Eqs. (7.39), (7.41) and (7.46), the dynamic structure factor, Eq. (7.34), becomes

$$S^{\alpha\beta}(\vec{k},\omega) = \frac{1}{2\pi}\left(\frac{e\hbar}{m_e c}\right)^2 |F(k)|^2 \sum_{j,\ell} e^{-i\vec{k}\cdot\vec{R}_j} e^{i\vec{k}\cdot\vec{R}_\ell}$$

$$\times \int d\tau\, e^{-i\omega\tau} \langle P^{\alpha}_{j}(\vec{k},0) P^{\beta}_{\ell}(\vec{k},\tau)\rangle \tag{7.49}$$

where, now, the target average,

$$\langle\cdots\rangle = \sum_{\Sigma_i} P_{\Sigma_i} \langle\Sigma_i| \cdots |\Sigma_i\rangle \tag{7.50}$$

is over initial target spin eigenstates, $|\Sigma_i\rangle$, of the Heisenberg Hamiltonian

$$H = - \sum_{j,\ell} J_{j,\ell} \vec{S}_j \cdot \vec{S}_\ell \tag{7.51}$$

which determines the time evolvement of $\vec{S}_\ell(\tau)$. The function F(k), which is given by

$$F(k) = \int d^3u \; e^{-i\vec{k}\cdot\vec{u}} \; \delta(\vec{u}) \tag{7.52}$$

i.e., the Fourier transform of the normalized spin density, is the magnetic atomic form factor. Notice, that since $\delta(\vec{u})$ is of microscopic range, F(k) will be a long-ranged, slowly varying function of $\vec{k}$. Also, the form factor is normalized to unity, i.e., F(0) = 1.

In the QA, which is applicable for critical scattering,

$$P_\ell^\beta(\vec{k},\tau) \cong P_\ell^\beta(\vec{k},0) \tag{7.53}$$

and Eq. (7.49) becomes

$$S^{\alpha\beta}(\vec{k},\omega) = \left(\frac{e\hbar}{m_e c}\right)^2 |F(k)|^2 \sum_{j,\ell} e^{-i\vec{k}\cdot\vec{R}_j} e^{i\vec{k}\cdot\vec{R}_\ell} \langle P_j^\alpha(\vec{k}) P_\ell^\beta(\vec{k}) \rangle \delta(\omega) \tag{7.54}$$

The neutron matrix elements, $M_N^{\alpha\beta}$, in Eq. (7.33) are evaluated for an unpolarized neutron beam. Summing over final neutron spin states and averaging over initial neutron spin states, from Eqs. (4.34)

and (7.31), one obtains

$$M_N^{\alpha\beta} = \frac{1}{2}\left(\frac{4\pi g e\hbar}{m_N c}\right)^2 \sum_{\sigma_i} \langle \sigma_i | s_N^\alpha s_N^\beta | \sigma_i \rangle = \frac{1}{4}\left(\frac{4\pi g e\hbar}{m_N c}\right)^2 \delta_{\alpha,\beta} \tag{7.55}$$

From Eqs. (7.54) and (7.55), the single scattering magnetic DDCS, Eq. (7.33) becomes

$$\frac{d^2\sigma(1)}{d\Omega dE_f} = \left(\frac{ge^2}{m_e c^2}\right)^2 \frac{1}{\hbar} |F(k)|^2 \sum_{j,\ell} e^{-i\vec{k}\cdot\vec{R}_j} e^{i\vec{k}\cdot\vec{R}_\ell} \langle P_j^\alpha(\vec{k}) P_\ell^\alpha(\vec{k}) \rangle \delta(\omega) \tag{7.56}$$

where the identity Eq. (5.8) has been utilized. From Eq. (7.47)

$$\begin{aligned} \langle P_j^\alpha(\vec{k}) P_\ell^\alpha(\vec{k}) \rangle &= \langle [\vec{S}_j - \hat{k}(\vec{S}_j \cdot \hat{k})] \cdot [\vec{S}_\ell - \hat{k}(\vec{S}_\ell \cdot \hat{k})] \rangle \\ &= \langle \vec{S}_j \cdot \vec{S}_\ell - (\vec{S}_j \cdot \hat{k})(\vec{S}_\ell \cdot \hat{k}) \rangle \\ &= (\delta_{\alpha,\beta} - \hat{k}^\alpha \hat{k}^\beta) \langle S_j^\alpha S_\ell^\beta \rangle \end{aligned} \tag{7.57}$$

where $\hat{k}^\alpha = k^\alpha/k$. For the Heisenberg Hamiltonian, Eq. (7.51), the target spin system is invariant to simultaneous rotations of the spins, i.e.,

$$\langle S_j^\alpha S_\ell^\beta \rangle = \frac{1}{3} \delta_{\alpha,\beta} \langle \vec{S}_j \cdot \vec{S}_\ell \rangle \tag{7.58}$$

thus, from Eq. (7.57),

$$\langle P_j^{\alpha}(\vec{k}) P_{\ell}^{\alpha}(\vec{k}) \rangle = \frac{2}{3} \langle \vec{S}_j \cdot \vec{S}_{\ell} \rangle \tag{7.59}$$

The single scattering magnetic DCS, from Eqs. (7.56) and (7.59), is

$$\frac{d\sigma^{(1)}}{d\Omega} = \frac{2}{3} \left(\frac{ge^2}{m_e c^2} \right)^2 |F(k)|^2 \sum_{j,\ell} e^{-i\vec{k}\cdot\vec{R}_j} e^{i\vec{k}\cdot\vec{R}_{\ell}} \langle \vec{S}_j \cdot \vec{S}_{\ell} \rangle \tag{7.60}$$

Assuming a translationally invariant spin system,

$$\sum_{j,\ell} e^{-i\vec{k}\cdot\vec{R}_j} e^{i\vec{k}\cdot\vec{R}_{\ell}} \langle \vec{S}_j \cdot \vec{S}_{\ell} \rangle = N \sum_{\ell} e^{i\vec{k}\cdot\vec{R}_{\ell}} \langle \vec{S}_0 \cdot \vec{S}_{\vec{R}_{\ell}} \rangle \tag{7.61}$$

As shown by Gersch, et al.[24] (also see Appendix V), one can write the sum in Eq. (7.61) as

$$\sum_{\ell} e^{i\vec{k}\cdot\vec{R}_{\ell}} \langle \vec{S}_0 \cdot \vec{S}_{\vec{R}_{\ell}} \rangle = \rho \sum_{\tau} \int d^3R \, e^{i(\vec{k}-\vec{\tau})\cdot\vec{R}} \gamma(\vec{R}) \tag{7.62}$$

where ρ (the inverse of the unit cell volume) is the atomic number density, since there is only one atom per unit cell, τ is a reciprocal lattice vector and

$$\gamma(\vec{R}) \equiv \langle \vec{S}_0 \cdot \vec{S}_{\vec{R}} \rangle. \tag{7.63}$$

From Eqs. (7.61) and (7.62), the single scattering magnetic DCS, Eq. (7.60), becomes

$$\frac{d\sigma}{d\Omega}^{(1)} = N \frac{2}{3} \left(\frac{ge^2}{m_e c^2} \right)^2 |F(k)|^2 \rho \sum_{\tau} \int d^3R \; e^{i(\vec{k}-\vec{\tau})\cdot\vec{R}} \gamma(\vec{R}) \qquad (7.64)$$

This DCS is now applied to critical magnetic scattering from a ferromagnet slightly above its Curie temperature.

Critical Magnetic Scattering

The Curie (critical) temperature, T_c, of a ferromagnet is defined as that temperature at which a ferromagnet spontaneously goes from its paramagnetic state to its ferromagnetic state. For the paramagnetic state ($T > T_c$), the Heisenberg spins, $\vec{S}_j$, in the absence of any external magnetic field, are randomly oriented, so that the mean total spin, $\langle \vec{S}_T \rangle$, is zero, i.e.,

$$\langle \vec{S}_T \rangle = \langle \sum_j \vec{S}_j \rangle = 0 \qquad \text{(paramagnetic, } T > T_c) \qquad (7.65)$$

In the ferromagnetic state ($T < T_c$), the spins are partially aligned and pointing in the same direction (within a given domain), thus

$$\langle \vec{S}_T \rangle \neq 0 \qquad \text{(ferromagnetic, } T < T_c) \qquad (7.66)$$

As the ferromagnet approaches its Curie temperature from above ($T \to T_c^+$), spin fluctuations become very large, i.e., the net spin of any given subregion of the magnet suffers large fluctuations about its mean value of zero. This is due to the fact that the magnet is approaching its ordered (ferromagnetic) state. These spin fluctuations

will magnetically scatter thermal-neutrons. Thus, the single scattering magnetic DCS, Eq. (7.64), will become very large as the Curie temperature is approached. This can be seen in a qualitative way by noting that the zero (external magnetic) field isothermal susceptibility, χ_T,

$$\chi_T \equiv \lim_{H \to 0} \left(\frac{\partial M}{\partial H}\right)_T \tag{7.67}$$

as determined from mean field theory, is given by[25]

$$\chi_T = \frac{A}{T - T_c} \qquad (T > T_c) \tag{7.68}$$

where A is a constant. In Eq. (7.67), M is the magnet's mean magnetic moment and H is an externally applied magnetic field. As shown in Appendix V, for a ferromagnet in its paramagnetic state ($T > T_c$),

$$T\chi_T = \frac{N g_e^2 \mu_B^2}{3k_B} \rho \sum_\tau \int d^3R \; e^{-i\vec{\tau}\cdot\vec{R}} \; \gamma(\vec{R}) \tag{7.69}$$

Therefore, by noting that when $\vec{k}$ is equal to a reciprocal lattice vector, $\vec{k} - \vec{\tau}$ is also equal to a reciprocal lattice vector, from Eqs. (7.64) and (7.69), it follows that

$$\left.\frac{d\sigma}{d\Omega}^{(1)}\right|_{\vec{k}=\vec{\tau}} \sim T\chi_T \tag{7.70}$$

and, from Eq. (7.68),

$$\left.\frac{d\sigma}{d\Omega}^{(1)}\right|_{\vec{k}=\vec{\tau}} \to \infty \qquad \text{as } T \to T_c \tag{7.71}$$

i.e., the scattering around the Bragg peaks of the crystal becomes huge near the Curie temperature. The form factor, F(k), in the DCS, Eq. (7.64), will serve to depress the magnitude of the scattering around all but the $\vec{\tau} = 0$ Bragg peak.

Considering only small-angle (small $\vec{k}$) scattering, i.e., scattering around $\vec{\tau} = 0$, the single scattering magnetic DCS, Eq. (7.64), can be written as

$$\frac{d\sigma}{d\Omega}^{(1)} = N \frac{2}{3}\left(\frac{ge^2}{m_e c^2}\right)^2 \rho \int d^3R \; e^{i\vec{k}\cdot\vec{R}} \gamma(\vec{R}) \tag{7.72}$$

where the form factor, F(k), has been set equal to unity since it is a slowly varying function of k and F(0) = 1.

The correlation of the fluctuations of the spin about its mean value, or the spin-spin correlation function, is given by

$$\langle\left[\vec{S}_0 - \langle\vec{S}_0\rangle\right] \cdot \left[\vec{S}_{\vec{R}} - \langle\vec{S}_{\vec{R}}\rangle\right]\rangle = \langle\vec{S}_0 \cdot \vec{S}_{\vec{R}}\rangle - \langle\vec{S}_0\rangle \cdot \langle\vec{S}_{\vec{R}}\rangle$$

$$= \langle\vec{S}_0 \cdot \vec{S}_{\vec{R}}\rangle - \langle S\rangle^2 \tag{7.73}$$

where

$$\vec{S} = \vec{S}_T/N \tag{7.74}$$

and the fact that $\langle \vec{S}_{\vec{R}} \rangle$ is independent of $\vec{R}$ has been utilized. Above T_c, from Eqs. (7.65) and (7.74), $\langle S \rangle^2 = 0$; thus, the spin-spin correlation function is equal to $\gamma(\vec{R})$, i.e.,

$$\gamma(\vec{R}) = \langle \vec{S}_0 \cdot \vec{S}_{\vec{R}} \rangle \tag{7.75}$$

When near T_c, the correlation function, $\gamma(\vec{R})$, can be determined utilizing the methods of Klein and Tisza, and Ornstein and Zernike as outlined in Chapter V. Letting $y_k = S_k^x + S_k^y + S_k^z$, from Eq. (5.28) the fluctuation, z_k, of y_k in the k^{th} "Klein-Tisza" cell is given by

$$z_k = \sum_\alpha \left(S_k^\alpha - \langle S_k^\alpha \rangle \right) = \sum_\alpha S_k^\alpha \tag{7.76}$$

Thus, for a translationally invariant spin system, the correlation function, $g_{0k} \equiv g_k$, from Eq. (5.33) is given by

$$g_k = \frac{\langle \sum_\alpha S_0^\alpha \sum_\beta S_k^\beta \rangle}{\left[\langle \left(\sum_\alpha S^\alpha \right)^2 \rangle \langle \left(\sum_\alpha S^\alpha \right)^2 \rangle \right]^{\frac{1}{2}}} \tag{7.77}$$

But

$$\left(\sum_\alpha S^\alpha \right)^2 = S^\alpha S^\alpha = S^2 \tag{7.78}$$

since, as is well-known, S^α and S^β $(\alpha \neq \beta)$ anticommute, i.e.,

$$S^\alpha S^\beta + S^\beta S^\alpha = 0 \qquad (\alpha \neq \beta) \tag{7.79}$$

and

$$\langle \sum_{\alpha} S_0^{\alpha} \sum_{\beta} S_k^{\beta} \rangle = \langle S_0^{\alpha} S_k^{\alpha} \rangle = \langle \vec{S}_0 \cdot \vec{S}_k \rangle \tag{7.80}$$

since, from Eq. (7.58),

$$\langle S_0^{\alpha} S_k^{\beta} \rangle = 0 \qquad (\alpha \neq \beta) \tag{7.81}$$

Therefore,

$$g_k = \frac{\langle \vec{S}_0 \cdot \vec{S}_k \rangle}{S(S+1)} \tag{7.82}$$

Letting the "Klein-Tisza" cell be equal in volume to the unit cell of the crystal, Eq. (7.82) becomes, for large R, from Eq. (5.52),

$$g(R) = \frac{\gamma(R)}{S(S+1)} = \frac{1}{4\pi\rho r_1^2} \frac{e^{-\kappa_1 R}}{R} \tag{7.83}$$

or

$$\gamma(R) = \frac{S(S+1)}{4\pi\rho r_1^2} \frac{e^{-\kappa_1 R}}{R} \tag{7.84}$$

The single scattering critical magnetic DCS for thermal-neutrons scattered from a ferromagnet slightly above its Curie temperature now becomes, from Eqs. (7.72) and (7.83),

$$\frac{d\sigma}{d\Omega}^{(1)} = \frac{N}{r_1^2}\frac{2}{3}S(S+1)\left(\frac{ge^2}{m_e c^2}\right)^2\left[\frac{1}{k^2+\kappa_1^2}\right] \tag{7.85}$$

Notice that the single scattering critical magnetic DCS has the same form as the single scattering critical nuclear DCS, Eq. (5.53), and can be obtained from it by the simple replacement of

$$a^2 \to \frac{2}{3}S(S+1)\left(\frac{ge^2}{m_e c^2}\right)^2 \tag{7.86}$$

In fact, as will be shown in Chapter VIII, with the appropriate approximations, the double scattering critical magnetic DCS can be obtained from the double scattering critical nuclear DCS, Eq. (6.37), by the same replacement given by (7.86).

The elastic nuclear scattering from ferromagnets occurs at the Bragg peaks. However, in practice, there is no problem, at least in the forward direction, in separating this scattering from the inelastic critical magnetic scattering, which also occurs around the Bragg peaks. For small-angle scattering, the elastic nuclear scattering is sharply peaked in the forward ($\vec{\tau} = 0$) direction. Experimentally, there is a small-angle limitation in measuring the critical magnetic cross section due to the presence of the forward neutron beam. This small-angle limitation will depend on the collimation of the neutron beam and other experimental factors. In an experiment on iron by Passell, et al.,[26] this small-angle limitation was approximately 1.5 degrees. The elastic nuclear scattering is only appreciable well within angles of this order; thus, it is wholly contained within the forward beam. This will also

be the case for elastic critical nuclear scattering from liquids, i.e., it will be wholly contained within the forward beam; thus, it was justifiably dropped from consideration in Chapters V and VI.

The elastic critical magnetic cross section is zero for ferromagnets above their Curie temperature, since $\langle \vec{S} \rangle = 0$ when $T > T_c$.

In the next chapter, the contribution of double scattering to the critical magnetic cross section is treated.

CHAPTER VIII

CRITICAL MAGNETIC DOUBLE SCATTERING FROM FERROMAGNETS

In this chapter, the double scattering cross section for critical magnetic scattering of thermal-neutrons from a ferromagnet slightly above its Curie temperature is obtained and compared to the single scattering cross section obtained in Chapter VII.

The decomposition of the four B operator correlation function appearing in the general double scattering DDCS, Eq. (4.36), into products of pair correlation functions, as given in Eq. (4.37), can be made for critical magnetic scattering for the same reasons as given in Chapter VI for critical nuclear scattering. For magnetic scattering, as was shown in Chapter VII, the operator B_ℓ, Eq. (6.2), is essentially the Heisenberg spin of the ℓ^{th} atom. For a ferromagnet slightly above its Curie temperature, the mean value of this spin is zero; thus there is no elastic magnetic scattering and the four B operator decomposition given by Eq. (4.37) becomes virtually exact.

The double scattering DDCS for magnetic scattering is given by Eq. (4.47),

$$\frac{d^2\sigma(2)}{d\Omega dE_f} = \lim_{\varepsilon\to 0} \frac{4m_N^4}{(2\pi\hbar)^8} \frac{1}{\hbar} \frac{k_f}{k_i} \int d^3k_a \int d^3k_b \int d\omega'$$

$$\times M_N^{\alpha\beta\gamma\delta} S^{\alpha\delta}(\vec{k}_i - \vec{k}_a, \vec{k}_b - \vec{k}_i, \omega') S^{\beta\gamma}(\vec{k}_a - \vec{k}_f, \vec{k}_f - \vec{k}_b, \omega - \omega')$$

$$\times \left(k_a^2 - k_i^2 + \frac{2m_N\omega'}{\hbar} + i\varepsilon\right)^{-1} \left(k_b^2 - k_i^2 + \frac{2m_N\omega'}{\hbar} - i\varepsilon\right)^{-1} \tag{8.1}$$

where the dynamic structure factor, $S^{\alpha\delta}(\vec{k}_2,\vec{k}_4,\omega')$, is given by Eq. (4.49),

$$S^{\alpha\delta}(\vec{k}_2,\vec{k}_4,\omega') = \frac{1}{2\pi}\int d\tau\ e^{-i\omega'\tau}\ \langle B^{\alpha}(\vec{k}_2,0)B^{\delta}(\vec{k}_4,\tau)\rangle \tag{8.2}$$

and the target operator, $B^{\alpha}(\vec{k}_2,\tau)$, is given by Eqs. (7.21) and (7.32),

$$B^{\alpha}(\vec{k}_2,\tau) = -\frac{e\hbar}{m_e c}\sum_j e^{-i\vec{k}_2\cdot\vec{r}_j(\tau)}\left\{\vec{s}_j(\tau) - \hat{k}_2\left[\vec{s}_j(\tau)\cdot\hat{k}_2\right]\right\}^{\alpha} \tag{8.3}$$

Assuming the ferromagnet to be made up of identical atoms with one atom per unit cell and neglecting nuclear thermal motions and electronic excitations, then, as shown in Chapter VII, by adopting the Heisenberg model for the electronic spin states, the dynamic structure factor can be written as

$$S^{\alpha\delta}(\vec{k}_2,\vec{k}_4,\omega') = \frac{1}{2\pi}\left(\frac{e\hbar}{m_e c}\right)^2 F(k_2)F(k_4)\sum_{j,n} e^{-i\vec{k}_2\cdot\vec{R}_j}\ e^{-i\vec{k}_4\cdot\vec{R}_n}$$

$$\times \int d\tau\ e^{-i\omega'\tau}\ \langle P_j^{\alpha}(k_2,0)P_n^{\delta}(k_4,\tau)\rangle \tag{8.4}$$

In Eq. (8.4), the target average is over initial target spin eigenstates of the Heisenberg Hamiltonian, Eq. (7.51), $F(k_2)$ is the magnetic atomic

form factor given by Eq. (7.52), the sums over j and n are over the atoms of the target and $P_j^{\alpha}(\vec{k}_2,\tau)$ is given by Eq. (7.47),

$$P_j^{\alpha}(\vec{k}_2,\tau) = \left\{ \vec{S}_j(\tau) - \hat{k}_2\left[\vec{S}_j(\tau) \cdot \hat{k}_2\right]\right\}^{\alpha} \tag{8.5}$$

where $\vec{S}_j$ is the effective Heisenberg spin operator of the j^{th} atom. In the QA, which is applicable for critical scattering,

$$S^{\alpha\delta}(\vec{k}_2,\vec{k}_4,\omega') = \left(\frac{e\hbar}{m_e c}\right)^2 F(k_2)F(k_4) \sum_{j,n} e^{-i\vec{k}_2\cdot\vec{R}_j} e^{-i\vec{k}_4\cdot\vec{R}_n}$$

$$\times \langle P_j^{\alpha}(\vec{k}_2)P_n^{\delta}(\vec{k}_4)\rangle\delta(\omega') \tag{8.6}$$

Similarly,

$$S^{\beta\gamma}(\vec{k}_1,\vec{k}_3,\tilde{\omega}) = \left(\frac{e\hbar}{m_e c}\right)^2 F(k_1)F(k_3) \sum_{\ell,m} e^{-i\vec{k}_1\cdot\vec{R}_\ell} e^{-i\vec{k}_3\cdot\vec{R}_m}$$

$$\times \langle P_\ell^{\beta}(\vec{k}_1)P_m^{\gamma}(\vec{k}_3)\rangle\delta(\tilde{\omega}) \tag{8.7}$$

The neutron spin matrix elements, $M_N^{\alpha\beta\gamma\delta}$, appearing in the DDCS, Eq. (8.1), are given by Eq. (4.48),

$$M_N^{\alpha\beta\gamma\delta} = \langle\sigma_i|a^{\alpha}a^{\beta}|\sigma_f\rangle\langle\sigma_f|a^{\gamma}a^{\delta}|\sigma_i\rangle \tag{8.8}$$

where $\vec{a}$ is given by Eq. (7.31),

$$\vec{a} = \frac{4\pi g e\hbar}{m_N c} \vec{s}_N \tag{8.9}$$

For an unpolarized neutron beam, summing over final neutron spin states and averaging over initial neutron spin states yields for these matrix elements

$$M_N^{\alpha\beta\gamma\delta} = \frac{1}{2}\left(\frac{4\pi g e\hbar}{m_N c}\right)^4 \sum_{\sigma_i} \langle \sigma_i | s_N^\alpha s_N^\beta s_N^\gamma s_N^\delta | \sigma_i \rangle \tag{8.10}$$

Utilizing the invariance of the Heisenberg Hamiltonian to simultaneous rotations of the spins, Eq. (7.58), a straight-forward calculation, given in Appendix VI, yields

$$M_N^{\alpha\beta\gamma\delta} \langle P_j^\alpha(\vec{k}_2) P_n^\delta(\vec{k}_4) \rangle \langle P_\ell^\beta(\vec{k}_1) P_m^\gamma(\vec{k}_3) \rangle$$

$$= \left(\frac{4\pi g e\hbar}{m_N c}\right)^4 f(\vec{k}_1, \vec{k}_2, \vec{k}_3, \vec{k}_4) \langle \vec{S}_j \cdot \vec{S}_n \rangle \langle \vec{S}_\ell \cdot \vec{S}_m \rangle \tag{8.11}$$

where

$$f = \frac{1}{144}\left\{\left[1+(\hat{k}_2\cdot\hat{k}_4)^2\right]\left[1+(\hat{k}_1\cdot\hat{k}_3)^2\right] + (\hat{k}_2\cdot\hat{k}_4)(\hat{k}_1\cdot\hat{k}_3)(\hat{k}_2\times\hat{k}_4)\cdot(\hat{k}_1\times\hat{k}_3)\right\} \tag{8.12}$$

From Eqs. (8.6), (8.7) and (8.11), the double scattering magnetic DDCS becomes

$$\frac{d^2\sigma(2)}{d\Omega dE_f} = 4\left(\frac{ge^2}{\pi m_e c^2}\right)\frac{1}{\hbar}\lim_{\varepsilon\to 0}\ \frac{k_f}{k_i}\int d^3k_a \int d^3k_b$$

$$\times f(\vec{k}_a-\vec{k}_f,\vec{k}_i-\vec{k}_a,\vec{k}_f-\vec{k}_b,\vec{k}_b-\vec{k}_i)F(|\vec{k}_i-\vec{k}_a|)F(|\vec{k}_a-\vec{k}_f|)F(|\vec{k}_f-\vec{k}_b|)F(|\vec{k}_b-\vec{k}_i|)$$

$$\times \sum_{j\ell mn} e^{-i(\vec{k}_i-\vec{k}_a)\cdot\vec{R}_j}\, e^{-i(\vec{k}_a-\vec{k}_f)\cdot\vec{R}_\ell}\, e^{-i(\vec{k}_f-\vec{k}_b)\cdot\vec{R}_m}\, e^{-i(\vec{k}_b-\vec{k}_i)\cdot\vec{R}_n}$$

$$\times \langle\vec{S}_j\cdot\vec{S}_n\rangle\langle\vec{S}_\ell\cdot\vec{S}_m\rangle(k_a^2-k_i^2+i\varepsilon)^{-1}(k_b^2-k_i^2-i\varepsilon)^{-1}\delta(\omega) \qquad (8.13)$$

At this point, the assumption that the spin system is translationally invariant will be made. As discussed in Chapter VI within the context of critical nuclear double scattering, this assumption implies a homogeneity of bulk effects and a neglection of boundary effects, yet a retention of the finite nature of the target which is crucial to multiple scattering. Assuming homogeneous bulk effects for the spin system implies that $\langle\vec{S}_j\cdot\vec{S}_n\rangle$ will depend only on the relative separation between spins, i.e.,

$$\langle\vec{S}_j\cdot\vec{S}_n\rangle = \langle\vec{S}_0\cdot\vec{S}_{\vec{R}_n-\vec{R}_j}\rangle = \langle\vec{S}_0\cdot\vec{S}_{\vec{R}_p}\rangle \qquad (8.14)$$

where $\vec{R}_p = \vec{R}_n - \vec{R}_j$. The sums over j and n in Eq. (8.13) now become, letting $\vec{R}_n = \vec{R}_p + \vec{R}_j$, then replacing $\vec{R}_p$ with $\vec{R}_n$,

$$\sum_{j,n} e^{-i(\vec{k}_i-\vec{k}_a)\cdot\vec{R}_j} e^{-i(\vec{k}_b-\vec{k}_i)\cdot\vec{R}_n} \langle\vec{S}_j\cdot\vec{S}_n\rangle$$

$$= \sum_{j,p} e^{-i(\vec{k}_i-\vec{k}_a)\cdot\vec{R}_j} e^{-i(\vec{k}_b-\vec{k}_i)\cdot(\vec{R}_p+\vec{R}_j)} \langle\vec{S}_0\cdot\vec{S}_{\vec{R}_p}\rangle$$

$$= \sum_{j,n} e^{i(\vec{k}_a-\vec{k}_b)\cdot\vec{R}_j} e^{-i(\vec{k}_b-\vec{k}_i)\cdot\vec{R}_n} \langle\vec{S}_0\cdot\vec{S}_{\vec{R}_n}\rangle \qquad (8.15)$$

Neglecting boundary effects implies that the j and n sums are independent of each other, and retaining the finite nature of the target implies that the j and n sums still go from one to N. Similarly, the sums over ℓ and m in Eq. (8.13) become, letting $\vec{R}_\ell = \vec{R}_m - \vec{R}_q$, then replacing $\vec{R}_m$ with $\vec{R}_\ell$ and $\vec{R}_q$ with $\vec{R}_m$,

$$\sum_{\ell,m} e^{-i(\vec{k}_a-\vec{k}_f)\cdot\vec{R}_\ell} e^{-i(\vec{k}_f-\vec{k}_b)\cdot\vec{R}_m} \langle\vec{S}_\ell\cdot\vec{S}_m\rangle$$

$$= \sum_{\ell,m} e^{-i(\vec{k}_a-\vec{k}_b)\cdot\vec{R}_\ell} e^{i(\vec{k}_a-\vec{k}_f)\cdot\vec{R}_m} \langle\vec{S}_0\cdot\vec{S}_{\vec{R}_m}\rangle \qquad (8.16)$$

Mathematically, the equality signs hold in Eqs. (8.15) and (8.16) only for a truly infinite target; in which case, for small-angle scattering, the sums over j and ℓ give zero unless $\vec{k}_a = \vec{k}_b$ and all target geometry, which is inherent in double scattering from finite targets, is lost.

However, for small-angle scattering from macroscopic targets, the sums over j and ℓ still give zero unless $|\vec{k}_a - \vec{k}_b| \lesssim \ell^{-1}$, where ℓ represents a linear dimension of the target. Thus, for thermal-neutrons

$$\vec{k}_a \cong \vec{k}_b \tag{8.17}$$

and Eqs. (8.15) and (8.16) represent physical equalities.

The integrations over $\vec{k}_a$ and $\vec{k}_b$ in Eq. (8.13) are now performed. Before doing so, however, Eq. (8.17) is used to simplify the factor f in Eq. (8.13), which is a much more slowly varying function of $\vec{k}_a$ and $\vec{k}_b$ than the exponential factors. Thus, from Eq. (8.12)

$$f(\vec{k}_a-\vec{k}_f,\vec{k}_i-\vec{k}_a,\vec{k}_f-\vec{k}_b,\vec{k}_b-\vec{k}_i) \cong f(\vec{k}_a-\vec{k}_f,\vec{k}_i-\vec{k}_a,\vec{k}_f-\vec{k}_a,\vec{k}_a-\vec{k}_i) = \frac{1}{36} \tag{8.18}$$

Using Eqs. (8.15), (8.16) and (8.18) and the explicit form for the form factor F, Eq. (7.52), the $\vec{k}_a$ and $\vec{k}_b$ integrations in Eq. (8.13) are easily evaluated using the methods of complex contour integration and yield for the double scattering magnetic DDCS,

$$\frac{d^2\sigma(2)}{d\Omega dE_f} = \frac{4}{9}\left(\frac{ge^2}{m_e c^2}\right)^4 \frac{1}{\hbar}\frac{k_f}{k_i} \int d^3u_1 \int d^3u_2 \int d^3u_3 \int d^3u_4$$

$$\times\, e^{-i\vec{k}_f\cdot\vec{u}_1}\, e^{i\vec{k}_i\cdot\vec{u}_2}\, e^{i\vec{k}_f\cdot\vec{u}_3}\, e^{-i\vec{k}_i\cdot\vec{u}_4}\, \delta(\vec{u}_1)\delta(\vec{u}_2)\delta(\vec{u}_3)\delta(\vec{u}_4)$$

$$\times \sum_{j\ell mn} e^{i\vec{k}_i\cdot\vec{R}_n} e^{-i\vec{k}_f\cdot\vec{R}_m} \frac{e^{-ik_i|\vec{R}_\ell-\vec{R}_j-\vec{R}_m-\vec{u}_1+\vec{u}_2|}}{|\vec{R}_\ell-\vec{R}_j-\vec{R}_m-\vec{u}_1+\vec{u}_2|}$$

$$\times \frac{e^{ik_i|\vec{R}_\ell-\vec{R}_j-\vec{R}_n-\vec{u}_3+\vec{u}_4|}}{|\vec{R}_\ell-\vec{R}_j-\vec{R}_n-\vec{u}_3+\vec{u}_4|} \langle\vec{S}_0\cdot\vec{S}_{\vec{R}_m}\rangle\langle\vec{S}_0\cdot\vec{S}_{\vec{R}_n}\rangle \tag{8.19}$$

Since $\vec{R}_m$ and $\vec{R}_n$ appear in the spin correlation functions, they are restricted to microscopic distances by the spin correlation range, κ_1^{-1}, i.e., $R_m \lesssim \kappa_1^{-1}$. Also, the $\vec{u}$ vectors are restricted by the normalized spin density functions, $\delta(\vec{u})$, which are of microscopic range. Therefore, since $\vec{R}_\ell$ and $\vec{R}_j$ are free to range over the entire target which is of macroscopic dimensions, the overwhelming contributions to the sums over j,ℓ come when

$$|\vec{R}_\ell-\vec{R}_j| \gg R_m, R_n, u \tag{8.20}$$

Thus,

$$|\vec{R}_\ell-\vec{R}_j-\vec{R}_m-\vec{u}_1+\vec{u}_2| \cong R_{\ell j}-\hat{R}_{\ell j}\cdot(\vec{R}_m+\vec{u}_1-\vec{u}_2) \tag{8.21}$$

$$|\vec{R}_\ell-\vec{R}_j-\vec{R}_n-\vec{u}_3+\vec{u}_4| \cong R_{\ell j}-\hat{R}_{\ell j}\cdot(\vec{R}_n+\vec{u}_3-\vec{u}_4) \tag{8.22}$$

where $\vec{R}_{\ell j} = \vec{R}_\ell - \vec{R}_j$ and $\hat{R}_{\ell j} = \vec{R}_{\ell j}/R_{\ell j}$. The exponential terms in Eq. (8.19) become

$$\frac{e^{-ik_i|\vec{R}_\ell-\vec{R}_j-\vec{R}_m-\vec{u}_1+\vec{u}_2|}}{|\vec{R}_\ell-\vec{R}_j-\vec{R}_m-\vec{u}_1+\vec{u}_2|} \simeq \frac{e^{-ik_i R_{\ell j}}\, e^{i\vec{q}\cdot(\vec{R}_m+\vec{u}_1-\vec{u}_2)}}{R_{\ell j}} \tag{8.23}$$

$$\frac{e^{ik_i|\vec{R}_\ell-\vec{R}_j-\vec{R}_n-\vec{u}_3+\vec{u}_4|}}{|\vec{R}_\ell-\vec{R}_j-\vec{R}_n-\vec{u}_3+\vec{u}_4|} \simeq \frac{e^{ik_i R_{\ell j}}\, e^{-i\vec{q}\cdot(\vec{R}_n+\vec{u}_3-\vec{u}_4)}}{R_{\ell j}} \tag{8.24}$$

where

$$\vec{q} = k_i \hat{R}_{\ell j} \tag{8.25}$$

represents the wave vector of magnitude k_i along the direction $\hat{R}_{\ell j}$. Identifying $\vec{R}_j$ as locating the first scattering event and $\vec{R}_\ell$ as locating the second scattering event, $\vec{q}$ is the intermediate wave vector of the neutron between scattering events.

Integrating Eq. (8.19) over E_f to obtain the DCS, using Eqs. (8.23) and (8.24) and letting the sums over j, ℓ, m and n go over to integrals, retaining only the $\vec{\tau} = 0$ reciprocal lattice vector for small-angle scattering, the double scattering magnetic DCS is

$$\frac{d\sigma}{d\Omega}^{(2)} = \frac{4}{9}\left(\frac{\rho g e^2}{m_c e^2}\right)^4 \int_V d^3R \int_{\langle\vec{R}\rangle} d^3R'\, R'^{-2}$$

$$\times\ F(|\vec{k}_i-\vec{q}|)|^2 |F(|\vec{q}-\vec{k}_f|)|^2 \hat{\gamma}(\vec{k}_i-\vec{q})\hat{\gamma}(\vec{q}-\vec{k}_f) \tag{8.26}$$

where

$$\hat{\gamma}(\vec{k}_1) = \int d^3r \; e^{i\vec{k}_1 \cdot \vec{r}} \; \gamma(r) \tag{8.27}$$

is the Fourier transform of $\gamma(r)$,

$$\gamma(r) = \langle \vec{S}_0 \cdot \vec{S}_{\vec{r}} \rangle \tag{8.28}$$

is the spin-spin correlation function and the change of integration variables $\vec{R}_j \to \vec{R}$, $\vec{R}_{\ell j} \to \vec{R}'$ has been made. The limits of integration on $\vec{R}'$ still go over the target but now depend on $\vec{R}$.

Using the OZ correlation function for $\gamma(r)$, Eq. (7.84), the double scattering critical magnetic DCS, Eq. (8.26) becomes

$$\frac{d\sigma^{(2)}}{d\Omega} = \frac{4}{9}\left[\frac{\rho S(S+1)}{r_1^2}\right]^2 \left(\frac{ge^2}{m_e c^2}\right)^4 \int_V d^3R \int_{\langle \vec{R} \rangle} d^3R' \; R'^{-2}$$

$$\times \; |F(|\vec{k}_i - \vec{q}|)|^2 |F(|\vec{q} - \vec{k}_f|)|^2 \left[|\vec{k}_i - \vec{q}|^2 + \kappa_1^2\right]^{-1} \left[|\vec{q} - \vec{k}_f|^2 + \kappa_1^2\right]^{-1} \tag{8.29}$$

Remembering that $\vec{q}$ depends on $\vec{R}'$, it becomes evident that the magnetic double scattering depends on target geometry. Now, using Eq. (8.25)

for $\vec{q}$, the magnitudes of the intermediate wave vector transfers are

$$|\vec{k}_i - \vec{q}| = 2k_i \sin \frac{\theta_1}{2} \tag{8.30}$$

$$|\vec{q} - \vec{k}_f| = 2k_i \sin \frac{\theta_2}{2} \tag{8.31}$$

where θ_1 and θ_2 are the intermediate scattering angles first introduced in Chapter VI. As the Curie temperature is approached, $\kappa_1 \to 0$ and the DCS, Eq. (8.29), is very sharply peaked around $\theta_1 = \theta_2 = 0$. Therefore, from Eqs. (8.30) and (8.31), the form factors appearing in the DCS, Eq. (8.29), are, to a good approximation,

$$F(|\vec{k}_i - \vec{q}|) \cong F(0) = 1 \tag{8.32}$$

$$F(|\vec{q} - \vec{k}_f|) \cong F(0) = 1 \tag{8.33}$$

since F is a slowly varying function of its argument and is normalized to unity. From Eqs. (8.32) and (8.33), the double scattering critical magnetic DCS, Eq. (8.29) becomes

$$\frac{d\sigma^{(2)}}{d\Omega} = \frac{\rho^2}{r_1^4} \left\{ \frac{2}{3} S(S+1) \left(\frac{ge^2}{m_e c^2} \right)^2 \right\}^2 \int_V d^3R \int_{\langle \vec{R} \rangle} d^3R' \, R'^{-2}$$

$$\times \left[|\vec{k}_i - \vec{q}|^2 + \kappa_1^2 \right]^{-1} \left[|\vec{q} - \vec{k}_f|^2 + \kappa_1^2 \right]^{-1} \tag{8.34}$$

This DCS can be obtained from the double scattering inelastic DCS for critical nuclear scattering of neutrons from liquids, Eqs. (6.31) and (6.32), by the simple replacement of the nuclear scattering length with the appropriate magnetic scattering length, i.e.,

$$a^2 \rightarrow \frac{2}{3} S(S+1) \left(\frac{ge^2}{m_e c^2} \right)^2 \tag{8.35}$$

Therefore, following the same analysis as presented in Chapter VI, from Eq. (6.37), the double scattering critical magnetic DCS is

$$\frac{d\sigma}{d\Omega}^{(2)} = \frac{N\rho \langle \ell \rangle}{r_1^4} \left\{ \frac{2}{3} S(S+1) \left(\frac{ge^2}{m_e c^2} \right)^2 \right\}^2 G(k_i, \theta_S, \kappa_1) \tag{8.36}$$

where $\langle \ell \rangle$ is given by Eq. (6.36) and G is given by Eqs. (6.38) - (6.40). The combined scattering cross section, single plus double, from Eqs. (7.85) and (8.36) is given by

$$\frac{d\sigma}{d\Omega} = \frac{d\sigma}{d\Omega}^{(1)} + \frac{d\sigma}{d\Omega}^{(2)} = \frac{d\sigma}{d\Omega}^{(1)} \left[1 + \beta H(x) \right] \tag{8.37}$$

where, now, the double scattering parameter β is given by

$$\beta = \frac{2\pi \langle \ell \rangle \rho S(S+1)}{3k_i^2 r_1^2} \left(\frac{ge^2}{m_e c^2} \right)^2 \tag{8.38}$$

and H(x) is given by Eq. (6.44).

In the next chapter, estimates of the critical magnetic double scattering contribution to the cross section are obtained for actual experiments performed.

CHAPTER IX

NUMERICAL RESULTS FOR CRITICAL MAGNETIC DOUBLE SCATTERING AND COMPARISON WITH FISHER'S MODIFICATION TO THE ORNSTEIN-ZERNIKE THEORY

In this chapter, numerical estimates are made for the double scattering contribution to the critical magnetic cross section using relevant parameters from actual critical scattering experiments performed on various ferromagnets. In addition, Fisher's modification[7] to the Ornstein-Zernike theory is considered and values for the critical exponent η are obtained which, when used in the first Born scattering approximation, reproduce the effect of double scattering.

Numerical Results for Critical Magnetic Double Scattering

Before any numerical results for critical magnetic double scattering are possible, the magnitude of the OZ "direct correlation length," r_1, must be determined. From Eq. (5.50), r_1 is given in terms of the second moment of the direct correlation function, a short-ranged function whose form depends on the Hamiltonian, Eq. (7.51). From mean-field theory, r_1 can be determined[27] and is given in terms of J(R), the exchange energy in Eq. (7.51),

$$r_1^2 = \frac{1}{6} \frac{\sum_{\vec{R}} R^2 J(\vec{R})}{\sum_{\vec{R}} J(\vec{R})} \tag{9.1}$$

Thus, considering nearest-neighbor interactions only,

$$r_1 \cong a_0/\sqrt{6} \tag{9.2}$$

where a_0 is the nearest-neighbor distance.

Using Eq. (9.2) and relevant parameters from an experiment on iron by Passell, et al.,[26] the double scattering critical magnetic DCS per target atom, obtained from Eq. (8.36), is plotted in Figure 2. Also plotted is the single scattering critical magnetic DCS per target atom, obtained from Eq. (7.85). Although, from this figure, the double scattering appears to be negligible, it does alter the Lorentzian line shape predicted by the first Born approximation.

In Table 1 are given theoretical values for the double scattering parameter β, Eq. (8.38), for actual critical magnetic scattering experiments performed on various ferromagnets.

Fisher's Modification to the Ornstein-Zernike Theory

As shown in Chapters V and VII, the OZ theory predicts that the correlation function, $\gamma(R)$, near the Curie temperature and for large R goes like

$$\gamma(R) \sim \frac{e^{-\kappa_1 R}}{R} \tag{9.3}$$

Therefore, the first Born or single scattering DCS, which depends on the Fourier transform of $\gamma(R)$, Eq. (7.72), goes like

$$\frac{d\sigma^{(1)}}{d\Omega} \sim \frac{1}{k^2 + \kappa_1^2} = \frac{1}{\kappa_1^2} \frac{1}{1 + x^2} \tag{9.4}$$

Thus, from Eq. (9.4), a plot of the inverse of the single scattering DCS versus x^2, an Ornstein-Zernike-Debye (OZD) plot, should be linear. However, experimental OZD plots for critical light scattering from liquids[28] display a small downward curvature for small x^2, thus indicating a non-Lorentzian line shape for the DCS. Fisher interpreted these experimental results as an indication of the failure of the OZ theory. He proposed a modification to the OZ theory by introducing a new critical exponent, η, in the correlation function, Eq. (9.3),

$$\tilde{\gamma}(R) = A(\eta) \frac{e^{-\kappa_1 R}}{R^{1+\eta}} \tag{9.5}$$

and estimated η to be less than 0.1, and probably $\eta \cong 0.05$. This η-modified correlation function, when used to obtain the single scattering DCS, does indeed produce the desired downward curvature for small x^2 in an OZD plot.

The possibility of multiple (double) scattering effects altering the Lorentzian line shape of the single scattering DCS, thus producing a non-linear OZD plot and giving rise to an apparent, double scattering induced η, has been recently investigated by Oxtoby and Gelbart[29] in connection with light scattering from liquids. An OZD plot of Eq. (8.37), which includes both singly and doubly scattered neutrons, for iron using the experimental parameters of Ref. 26 is shown in Figure 3,

and a definite downward curvature is noticeable. This curvature is difficult to detect in experimental neutron work in ferromagnets owing to the small-angle limitations discussed in Chapter VII. For example, the minimum value for x^2 in Figure 3 obtained by Passell, et al., corresponds to $x^2 \cong 4.5$.

To obtain values of an apparent η due to double scattering effects, the coefficient $A(\eta)$ in Eq. (9.5) is determined by requiring that

$$\int d^3R\ \tilde{\gamma}(R) = \int d^3R\ \gamma(R) \tag{9.6}$$

where $\gamma(R)$ is given by Eq. (7.84). This yields for $A(\eta)$,

$$A(\eta) = \frac{S(S+1)}{4\pi\rho r_1^2}\ \frac{\kappa_1^{-\eta}}{\Gamma(2-\eta)} \tag{9.7}$$

where Γ is the familiar gamma function.[30] Inserting the η-modified correlation function, $\tilde{\gamma}$, Eq. (9.5), into the single scattering DCS, Eq. (7.72), gives the η-modified single scattering DCS[31]

$$\frac{d\tilde{\sigma}}{d\Omega} = D\ \frac{\Gamma(1-\eta)}{\Gamma(2-\eta)}\ \frac{\sin[(1-\eta)\ \tan^{-1}x]}{x\left[1 + x^2\right]^{\frac{1-\eta}{2}}} \tag{9.8}$$

where

$$D = \frac{N}{r_1^2}\frac{2}{3}S(S+1)\left(\frac{ge^2}{m_e c^2}\right)^2\frac{1}{\kappa_1^2} \tag{9.9}$$

Neglecting the η dependence in the gamma functions and in the sine function yields

$$\frac{\hat{d\sigma}}{d\Omega} = \frac{D}{[1+x^2]^{1-\eta/2}} \tag{9.10}$$

since

$$\Gamma(1) = \Gamma(2) = 1 \tag{9.11}$$

and

$$\sin[\tan^{-1}x] = \frac{x}{(1+x^2)^{\frac{1}{2}}} \tag{9.12}$$

Equation (9.10) is the familiar form for the η-modified DCS first given by Fisher.

Returning to Eq. (9.8) and assuming that η is small, $\frac{\hat{d\sigma}}{d\Omega}$ can be expanded in terms of powers of η by noting that

$$\Gamma(2-\eta) = (1-\eta)\Gamma(1-\eta) \tag{9.13}$$

since

$$\Gamma(1+z) = z\Gamma(z). \tag{9.14}$$

Therefore,

$$\frac{\Gamma(1-\eta)}{\Gamma(2-\eta)} = \frac{1}{1-\eta} = 1 + \eta + \eta^2 + \ldots \tag{9.15}$$

Also,

$$\sin\left[(1-\eta)\tan^{-1}x\right] = \sin\left[\tan^{-1}x\right]\cos\left[\eta\tan^{-1}x\right]$$

$$- \sin\left[\eta\tan^{-1}x\right]\cos\left[\tan^{-1}x\right]$$

$$= \frac{x}{(1+x^2)^{\frac{1}{2}}}\left\{1 - \eta\,\frac{\tan^{-1}x}{x} - \eta^2\,\frac{(\tan^{-1}x)^2}{2} + \ldots\right\} \tag{9.16}$$

and

$$\frac{1}{\left[1+x^2\right]^{-\frac{\eta}{2}}} = e^{\frac{\eta}{2}\,\ell n(1+x^2)}$$

$$= 1 + \frac{\eta}{2}\,\ell n(1+x^2) + \frac{1}{2}\left[\frac{\eta}{2}\,\ell n(1+x^2)\right]^2 + \ldots \tag{9.17}$$

From Eqs. (9.15), (9.16) and (9.17), and retaining only terms linear in η, the η-modified DCS, Eq. (9.8), becomes

$$\frac{\tilde{d\sigma}}{d\Omega} = \frac{d\sigma}{d\Omega}^{(1)} [1 + \eta J(x)] \tag{9.18}$$

where $\frac{d\sigma}{d\Omega}^{(1)}$ is given by Eq. (7.85) and

$$J(x) = 1 - x^{-1}\tan^{-1}x + \frac{1}{2}\ell n(1+x^2) \tag{9.19}$$

Now, equating Eq. (9.18) to the DCS containing both single and double scattering, Eq. (8.37), a double scattering induced η is obtained,

$$\eta = K(x)\beta \tag{9.20}$$

where

$$K(x) \equiv \frac{H(x)}{J(x)} , \tag{9.21}$$

H(x) given by Eq. (6.44). The function K(x) is plotted in Figure 4. As $T \to T_c$, for a fixed, non-zero scattering angle, $x \to \infty$. In this limit,

$$\eta = 4\beta \qquad (x \to \infty) \tag{9.22}$$

Therefore, for an experiment performed in this region, an apparent η, induced from double scattering, of 0.05 or greater would result for experimental conditions where $\beta \geq 0.0125$. The function K(x) has a

minimum of 2.5 at $x \stackrel{\sim}{=} 2$; therefore, this would be the optimal region to probe experimentally in search for a real η. However, even at $x \stackrel{\sim}{=} 2$, experiments for which $\beta \geq 0.02$ will produce an apparent $\eta \geq 0.05$.

In Table 1 are given the values for η experimentally measured in critical magnetic scattering experiments performed on various ferromagnets. Comparing these values for η with the corresponding values for β, from Eq. (9.20) it becomes evident that the deviations in Lorentzian line shape produced by double scattering can compete with similar line shape deviations predicted by the η-modified OZ correlation function.

In the next chapter, it is shown that by restricting the range of applicability of the formalism in Chapters III and IV from the outset to critical scattering, Glauber's high energy approximation can be used to provide a much simpler and more powerful description of multiple scattering effects.

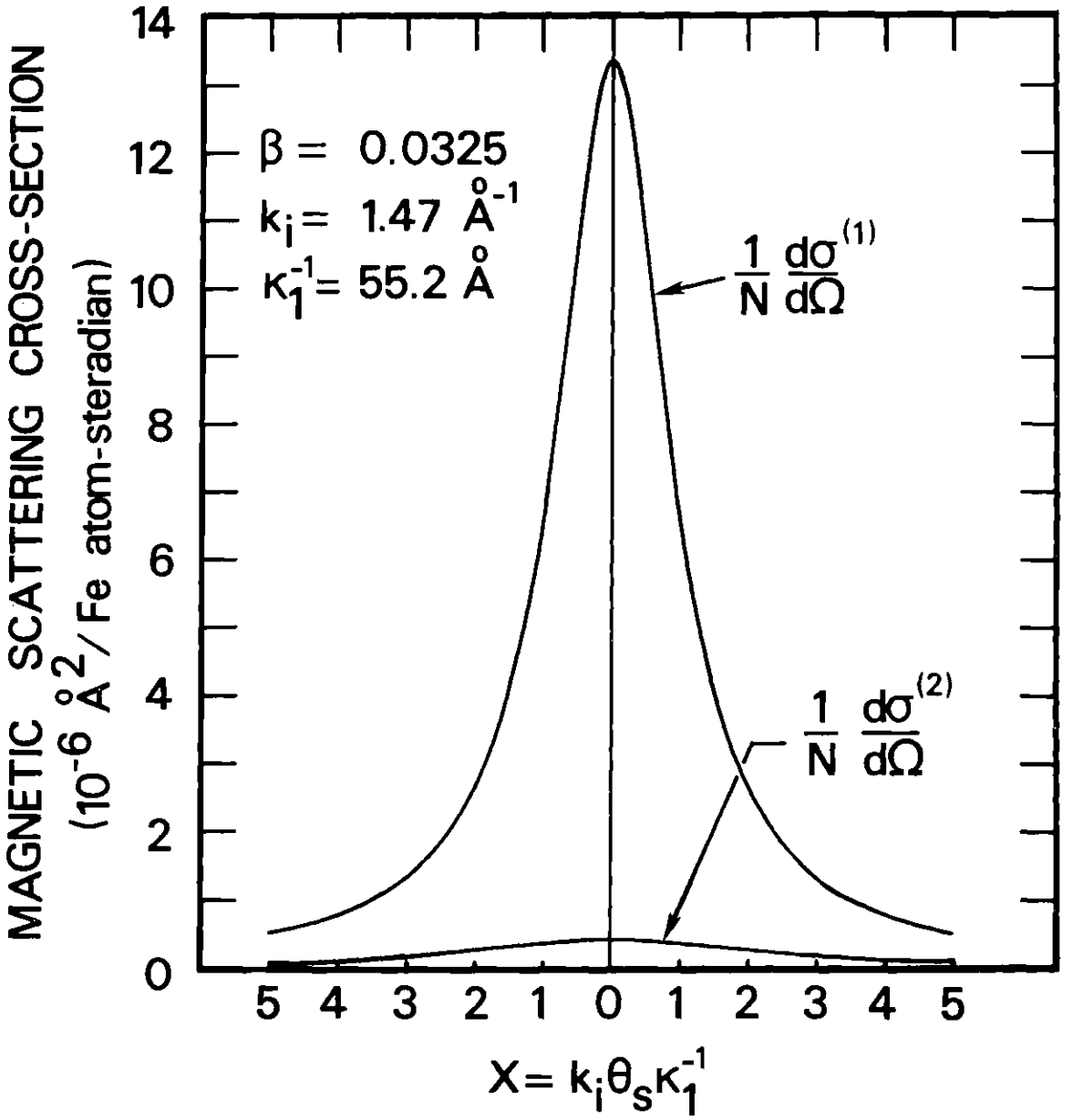

Figure 2. Theoretical Single and Double Scattering Cross Sections Using Relevant Parameters from an Experiment on Fe by Passell, et al. (Experimentally probed region corresponds to $2.1 \lesssim x \lesssim 3.9$.)

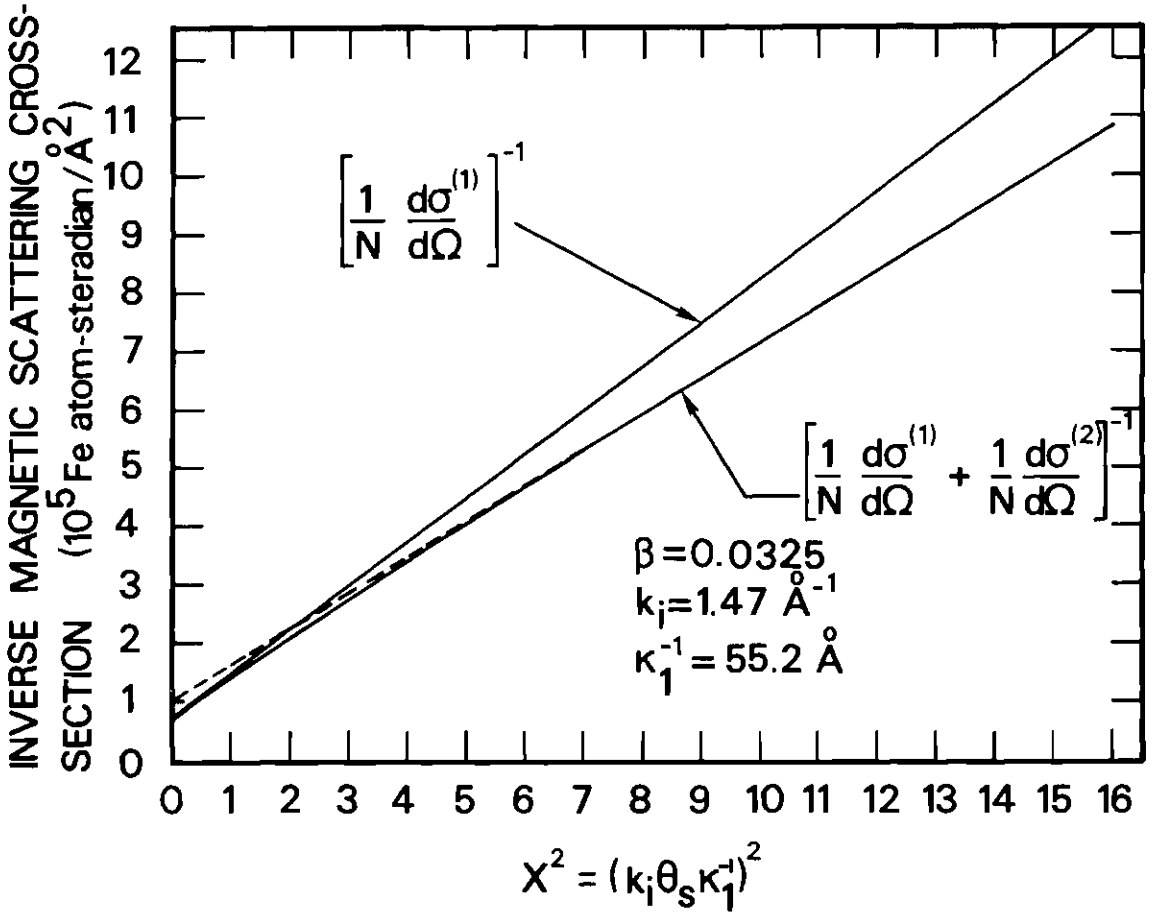

Figure 3. Theoretical OZD Plots of Single Scattering and Single-Plus-Double Scattering Cross Sections Using Relevant Parameters from an Experiment on Fe by Passell, et al. (Experimentally probed region corresponds to $4.5 \lesssim x^2 \lesssim 15.2$.)

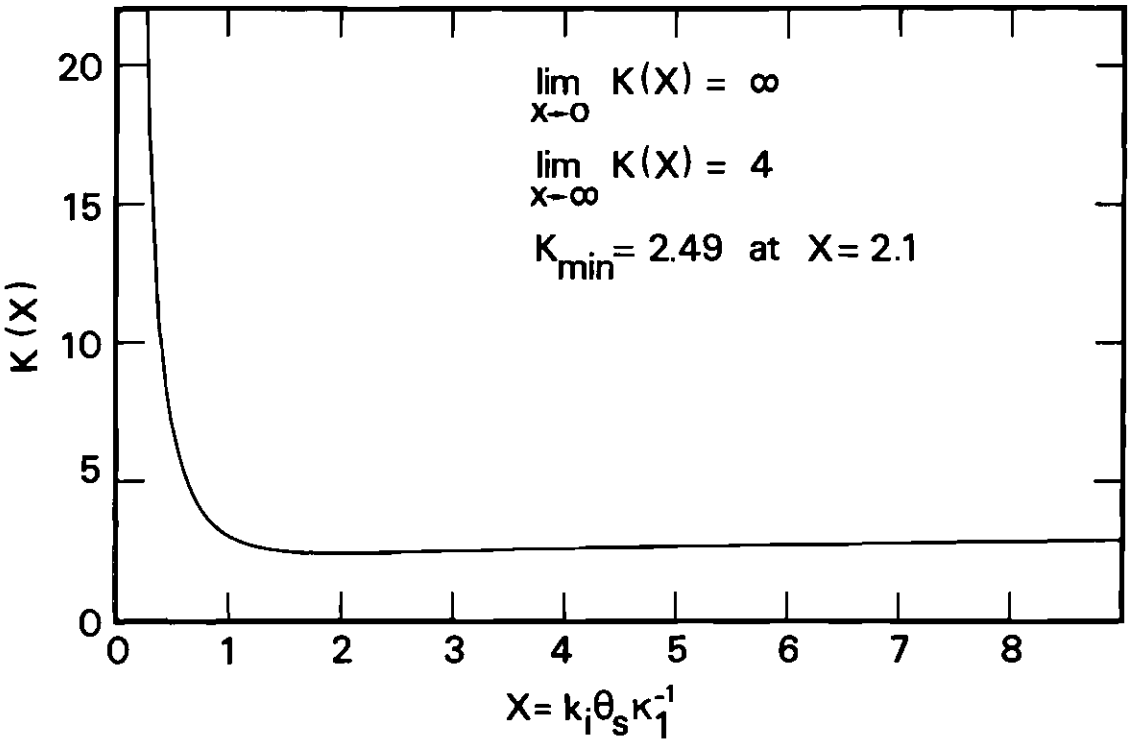

Figure 4. Function K(x) Appearing in Eq. (9.21) Versus x.

Table 1. Experimental Parameters and Theoretical β Values for Several Experiments on Magnetic Scattering of Neutrons from Ferromagnets.

Sample	k_i (Å^{-1})	r_1 (Å)[a]	Target size: Thickness (mm)	Experimental range of x	β	Experimental η
Fe[b]	1.47	1.01	12	$0.35 \lesssim x \lesssim 3.9$	0.032	c
			4.3		0.012	c
Fe[d]	5.03	1.01	2.5	$0.2 \lesssim x \lesssim 4.0$	0.00058	0.07 ± 0.05
EuO[e]	4.30	1.48	<1	$0.1 \lesssim x \lesssim 5.4$	0.00035	$-0.10 \lesssim \eta \lesssim 0.06$[f]
EuS[e]	4.30	1.72	<1	$0.35 \lesssim x \lesssim 3.5$	0.00017	$-0.07 \lesssim \eta \lesssim 0.11$[f]

a Theoretically determined from Eq. (9.2).

b Reference 26.

c Curves with $\eta = 0.10$, 0.15 and 0.20 fitted experimental curve about as well as curve with $\eta = 0$.

d Reference 32.

e Reference 33.

f η determined by scaling relation: $\eta = 2 - \frac{\gamma}{\nu}$.

CHAPTER X

THERMAL-NEUTRON CRITICAL MULTIPLE SCATTERING VIA THE GLAUBER APPROXIMATION

In this chapter, it is shown that by restricting the range of applicability of the multiple scattering theory presented in Chapters III and IV from the outset to critical scattering, Glauber's high energy approximation[8] can be used to provide a much simpler and more powerful description of multiple scattering effects.

The Glauber Approximation

The Glauber high energy approximation (GA) is applicable for those scattering conditions where the energy of the incident probe particle, E_i, is much greater than the absolute magnitude of the interaction, $|\phi|$, i.e.,

$$\frac{|\phi|}{E_i} <<< 1 \tag{10.1}$$

and the range of the interaction, ξ, is much greater than the magnitude of the inverse wave vector of the incident probe particle, k_i^{-1}, i.e.,

$$k_i \xi >>> 1 \tag{10.2}$$

From condition (10.1), one would expect that the probe particle's energy

would change very little in the scattering process, i.e., the scattering is predominantly quasielastic. Also, considering the uncertainty principle, i.e.,

$$k\xi \sim 1 \tag{10.3}$$

where k is the probe's wave vector transfer, i.e., $k = 2k_i \sin \frac{\theta_S}{2}$ (since the scattering is quasielastic), from condition (10.2), one would expect the scattering to be highly peaked in the forward direction, since from (10.2) and (10.3),

$$\sin \frac{\theta_S}{2} \sim \frac{1}{2k_i\xi} \ll 1 \tag{10.4}$$

These two conditions necessary for the applicability of the GA, i.e., quasielastic and small-angle scattering, are well satisfied for thermal-neutron critical scattering. Thus, it appears that the GA is applicable to thermal-neutron critical scattering.

That this is the case may not be obvious when applying conditions (10.1) and (10.2) directly to thermal-neutron critical scattering. For example, one might expect that condition (10.1) would not be satisfied for thermal-neutrons, due to their low energy content, $E_i \sim 10^{-3} - 10^{-1}$ eV and, that condition (10.2) would not be satisfied for nuclear scattering of thermal-neutrons, since the interaction, the Fermi-pseudo potential, essentially has a range of zero. However, the thermal-neutron scattering considered here is from macroscopic targets and the neutron

"feels" the effects of many target atoms (regardless of whether it scatters only once or undergoes multiple scatterings). Therefore, the interaction between the neutron and one target atom is not what is important in testing the validity of conditions (10.1) and (10.2), but rather the "effective interaction" between the neutron and the target as a whole. That is, replacing the many-particle macroscopic target with a fixed scattering center, the effective interaction is that interaction associated with the fixed scattering center that exactly reproduces the scattering from the macroscopic target. To determine this effective interaction, or optical model potential, only the first Born approximation need be considered. For an interaction that depends on distance only, the first Born DCS for a neutron scattering from a fixed scattering center with an interaction ϕ is, from Eq. (4.26) with $S(\vec{k},\omega) = \delta(\omega)$,

$$\frac{d\sigma}{d\Omega}^{(1)} = \left(\frac{m_N}{2\pi\hbar^2}\right)^2 \int d^3r \int d^3r' \, e^{-i\vec{k}\cdot(\vec{r}-\vec{r}')} \, \phi(\vec{r})\phi(\vec{r}') \qquad (10.5)$$

For thermal-neutron critical nuclear inelastic single scattering, the DCS is, from Eq. (5.15),

$$\left(\frac{d\sigma}{d\Omega}\right)_i^{(1)} = \left(\frac{m_N}{2\pi\hbar^2}\right)^2 \int d^3r \int d^3r' \, e^{-i\vec{k}\cdot(\vec{r}-\vec{r}')} \left(\frac{2\pi\hbar^2 a}{m_N}\right)^2 \Gamma(\vec{r},\vec{r}') \qquad (10.6)$$

where $\Gamma(\vec{r},\vec{r}')$ follows from Eqs. (5.23) and (5.52), dropping the self-

correlation term,

$$\Gamma(\vec{r},\vec{r}') = \frac{\rho}{4\pi r_1^2} \frac{e^{-\kappa_1|\vec{r}-\vec{r}'|}}{|\vec{r}-\vec{r}'|} \tag{10.7}$$

To obtain an estimate of the absolute magnitude of Γ, it is evaluated at $|\vec{r}-\vec{r}'| = \kappa_1^{-1}$. Thus, comparing Eqs. (10.5) and (10.6), an estimate of the magnitude of the effective interaction for thermal-neutron critical nuclear scattering can be obtained,

$$|\phi| \cong \frac{2\pi\hbar^2 a}{m_N} \left(\frac{\rho e^{-1}}{4\pi r_1^2 \kappa_1^{-1}}\right)^{1/2} \sim \left(\frac{\hbar^2 a \rho^{1/2}}{m_N r_1}\right) \kappa_1^{1/2} \tag{10.8}$$

For typical values of a, ρ and r_1,

$$|\phi| \sim 10^{-8}\ \kappa_1^{1/2}\ \text{eV} \tag{10.9}$$

where κ_1 is measured in inverse Angstroms. Thus, condition (10.1) is well satisfied for thermal-neutron critical scattering. The range of the effective interaction is obviously

$$\xi \sim \kappa_1^{-1} \tag{10.10}$$

so that condition (10.2) is well satisfied for thermal-neutron critical scattering if the target is sufficiently close to its critical point,

since, for thermal-neutrons, $k_i \sim 1\ \mathring{A}^{-1}$. Notice that condition (10.2) is almost never satisfied experimentally for critical light scattering, since for light $k_i \sim 10^{-3}\ \mathring{A}^{-1}$. The final justification for using the GA to describe thermal-neutron critical scattering is, of course, obtaining results that are in agreement with those obtained previously from first principles.

The details of the GA are presented in Ref. 8 and will not be treated here. Essentially, the GA to the scattering cross section is a quasielastic approximation in which, when treating the phase shifts (in the wave representing the probe) induced by the scattering, the probe is assumed to have traversed the target along a straight line parallel to $\vec{k}_i$. In the GA, the scattering amplitude, $F_{fi}(\vec{k}_f,\vec{k}_i)$, for a neutron to scatter from an initial plane wave state, $|\vec{k}_i\rangle$, to a final plane wave state, $|\vec{k}_f\rangle$, while the target goes from an initial state, $|T_i\rangle$, to a final state, $|T_f\rangle$, Eq. (172) of Ref. 8, is given by

$$F_{fi}(\vec{k}_f,\vec{k}_i) = \frac{k_i}{2\pi i}\int_{-\infty}^{\infty} d^2b\ e^{i\vec{k}\cdot\vec{b}}\ \langle T_f|e^{i\chi(\vec{b})} - 1|T_i\rangle \qquad (10.11)$$

where $\vec{b}$ is the impact parameter lying in a plane (chosen to be the x-y plane) perpendicular to $\vec{k}_i$. The phase shift $\chi(\vec{b})$ is the sum of the phase shifts induced by each of the N target atoms,

$$\chi(\vec{b}) = -\frac{m_N}{\hbar^2 k_i}\int_{-\infty}^{\infty} dz \sum_{j=1}^{N} \phi(\vec{b} + \hat{k}_i z - \vec{r}_j) \qquad (10.12)$$

The position vector of the neutron is given by $\vec{b} + \hat{k}_i z$ and $\vec{r}_j$ is the position vector of the j^{th} target atom.[34] Here, only an interaction that depends on distance is considered.

The DCS is obtained from Eq. (10.11) by multiplying $F_{fi}(\vec{k}_f, \vec{k}_i)$ by its complex conjugate, summing over final target states and averaging over initial target states,

$$\frac{d\sigma}{d\Omega} = \left(\frac{k_i}{2\pi}\right)^2 \int d^2b \int d^2b' \, e^{i\vec{k}\cdot(\vec{b}-\vec{b}')}$$

$$\times \left\langle \left(e^{i\chi(\vec{b})} - 1\right)\left(e^{-i\chi(\vec{b}')} - 1\right)\right\rangle \qquad (10.13)$$

The DCS given by Eq. (10.13) can be separated into a purely elastic part and an inelastic part,

$$\left(\frac{d\sigma}{d\Omega}\right) = \left(\frac{d\sigma}{d\Omega}\right)_e + \left(\frac{d\sigma}{d\Omega}\right)_i \qquad (10.14)$$

where

$$\left(\frac{d\sigma}{d\Omega}\right)_e = \left|\frac{k_i}{2\pi i} \int d^2b \, e^{i\vec{k}\cdot\vec{b}} \left(1 - \langle e^{i\chi(\vec{b})}\rangle\right)\right|^2 \qquad (10.15)$$

and

$$\left(\frac{d\sigma}{d\Omega}\right)_i = \left(\frac{k_i}{2\pi}\right)^2 \int d^2b \int d^2b' \, e^{i\vec{k}\cdot(\vec{b}-\vec{b}')}$$

$$\times \left\{\langle e^{i\chi(\vec{b})} \, e^{-i\chi(\vec{b}')}\rangle - \langle e^{i\chi(\vec{b})}\rangle\langle e^{-i\chi(\vec{b}')}\rangle\right\} \qquad (10.16)$$

For a macroscopic liquid target, the elastic DCS is non-negligible only in the forward direction, and for a macroscopic crystal, the elastic DCS is non-negligible only when $\vec{k}$ equals a reciprocal lattice vector of the crystal. The elastic DCS will now be dropped from further considerations.

For critical scattering, as shown in Appendix II, the free energy of the target can be adequately expressed as a quadratic form in terms of density fluctuations for nuclear scattering, or spin fluctuations for magnetic scattering. Thus, as a consequence, correlations of these fluctuations, to all orders, decompose into products of, at most, pair correlations. Therefore, it would be natural to expand the target averages in Eq. (10.16) in a cumulant expansion, since the cumulants are expressed in terms of correlations of fluctuations. Carrying the expansion out to the second cumulant includes products of, at most, pair correlations to all orders. As a consequence, truncation of the cumulant expansion at the second cumulant becomes virtually exact for critical scattering.

The cumulants, $\{w_n\}$, are defined by

$$\langle e^{i\lambda\chi(\vec{b})} \rangle = e^{\sum_{n=1}^{\infty} \lambda^n w_n} \tag{10.17}$$

where λ is set equal to unity once the cumulants have been determined. Differentiating Eq. (10.17) with respect to λ, then setting λ equal to zero yields

$$w_1 = i\langle\chi(\vec{b})\rangle \tag{10.18}$$

Differentiating Eq. (10.17) twice with respect to λ, then setting λ equal to zero yields

$$2w_2 + w_1^2 = -\langle\chi^2(\vec{b})\rangle \tag{10.19}$$

or

$$w_2 = -\frac{1}{2}\left[\langle\chi^2(\vec{b})\rangle - \langle\chi(\vec{b})\rangle^2\right] \tag{10.20}$$

Therefore, truncating the expansion at the second cumulant, from Eq. (10.17), setting λ equal to unity,

$$\langle e^{i\chi(\vec{b})}\rangle = e^{w_1+w_2} = e^{i\langle\chi(\vec{b})\rangle}\, e^{-\frac{1}{2}\left[\langle\chi^2(\vec{b})\rangle - \langle\chi(\vec{b})\rangle^2\right]}$$

$$\equiv e^{i\chi_{opt}(\vec{b})} \tag{10.21}$$

Similarly,

$$\langle e^{-i\chi(\vec{b}')}\rangle = e^{-i\chi^*_{opt}(\vec{b}')} \tag{10.22}$$

and

$$\langle e^{i\chi(\vec{b})}\ e^{-i\chi(\vec{b}')}\rangle =$$

$$e^{i\chi_{opt}(\vec{b})}\ e^{-i\chi^{*}_{opt}(\vec{b}')}\ e^{\langle\chi(\vec{b})\chi(\vec{b}')\rangle - \langle\chi(\vec{b})\rangle\langle\chi(\vec{b}')\rangle} \tag{10.23}$$

Thus, the inelastic DCS, Eq. (10.16), becomes

$$\left(\frac{d\sigma}{d\Omega}\right)_i = \left(\frac{k_i}{2\pi}\right)^2 \int d^2b \int d^2b'\ e^{i\vec{k}\cdot(\vec{b}-\vec{b}')}\ e^{i\chi_{opt}(\vec{b})}\ e^{-i\chi^{*}_{opt}(\vec{b}')}$$

$$\times \left\{ e^{\langle\chi(\vec{b})\chi(\vec{b}')\rangle - \langle\chi(\vec{b})\rangle\langle\chi(\vec{b}')\rangle} - 1 \right\} \tag{10.24}$$

The various terms appearing in the cross section, Eq. (10.24), have the following physical interpretation. The real part of $\chi_{opt}(\vec{b})$,

$$\text{Re}\ \chi_{opt}(\vec{b}) = \langle\chi(\vec{b})\rangle \tag{10.25}$$

corresponds to a real effective optical potential, or an index of refraction term. Its imaginary part,

$$\text{Im}\ \chi_{opt}(\vec{b}) = \frac{1}{2}\left[\langle\chi^2(\vec{b})\rangle - \langle\chi(\vec{b})\rangle^2\right] \tag{10.26}$$

represents a depletion, or extinction, of the neutron beam due to the inelastic scattering. Finally, the term in brackets represents the inelastic scattering, to all orders.

At this point, the inelastic DCS given by Eq. (10.24) is compared to the perturbative expansion of the DDCS, Eq. (4.1), in Chapter IV in order to justify the physical interpretation given for the neglected terms in Δ_3 and Δ_4. Although these two cross sections are vastly different in form, they still can be compared by realizing that, at least in principle, the above DCS should be obtainable from the DDCS in Chapter IV, using the appropriate approximations. Thus, noting that the perturbative expansion of the DDCS in Chapter IV is in terms of powers of the interaction, $\phi = AB$, a one to one correspondence can be made with the above DCS by expanding it in terms of powers of the interaction ϕ, or equivalently, from Eq. (10.12), in terms of powers of χ. Therefore, from Eq. (10.24),

$$\left(\frac{d\sigma}{d\Omega}\right)_i = \left(\frac{k_i}{2\pi}\right)^2 \int d^2b \int d^2b' \, e^{i\vec{k}\cdot(\vec{b}-\vec{b}')} \sum_{n=2}^{\infty} \delta_n \tag{10.27}$$

where, the first three terms are given by

$$\delta_2 = \langle\chi(\vec{b})\chi(\vec{b}')\rangle - \langle\chi(\vec{b})\rangle\langle\chi(\vec{b}')\rangle \tag{10.28}$$

$$\delta_3 = i \, \mathrm{Re} \, \chi_{opt}(\vec{b})[\langle\chi(\vec{b})\chi(\vec{b}')\rangle - \langle\chi(\vec{b})\rangle\langle\chi(\vec{b}')\rangle]$$

$$- i \, \mathrm{Re} \, \chi^*_{opt}(\vec{b}')[\langle\chi(\vec{b})\chi(\vec{b}')\rangle - \langle\chi(\vec{b})\rangle\langle\chi(\vec{b}')\rangle] \tag{10.29}$$

$$\delta_4 = -\left\{\text{Im } \chi_{opt}(\vec{b}) - \frac{1}{2}\left[\text{Re } \chi_{opt}(\vec{b})\right]^2\right\}$$

$$\times \left[\langle\chi(\vec{b})\chi(\vec{b}')\rangle - \langle\chi(\vec{b})\rangle\langle\chi(\vec{b}')\rangle\right]$$

$$+ \frac{1}{2}\left[\langle\chi(\vec{b})\chi(\vec{b}')\rangle - \langle\chi(\vec{b})\rangle\langle\chi(\vec{b}')\rangle\right]^2$$

$$- \left\{\text{Im } \chi^*_{opt}(\vec{b}') - \frac{1}{2}\left[\text{Re } \chi^*_{opt}(\vec{b}')\right]^2\right\}$$

$$\times \left[\langle\chi(\vec{b})\chi(\vec{b}')\rangle - \langle\chi(\vec{b})\rangle\langle\chi(\vec{b}')\rangle\right] \qquad (10.30)$$

A one to one correspondence between δ_2, δ_3 and δ_4 given above can be made with Δ_2, Δ_3 and Δ_4 given by Eqs. (4.8) - (4.10). Notice that the 1,2 notation of the latter has gone over, respectively, into the b,b′ dependence of the former. Clearly, as stated in Chapter IV, δ_2 (or Δ_2) represents single scattering, δ_3 (or Δ_3) represents the mixed effects of refraction and single scattering, the first and last terms of δ_4 (or Δ_4) represent the mixed effects of extinction, refraction and single scattering and the middle term in δ_4 (or Δ_4) represents double scattering.

Critical Nuclear Scattering

The formalism just presented is now applied to the case of critical nuclear scattering of thermal-neutrons from liquids. The interaction, ϕ, which determines the phase shift $\chi(\vec{b})$, Eq. (10.12), is given by the Fermi-pseudo potential, Eq. (5.1). Thus, from Eq. (10.12), the target average of $\chi(\vec{b})$ is then

$$\langle \chi(\vec{b}) \rangle = - \frac{2\pi a}{k_i} \int dz \langle n(\vec{b},z) \rangle \tag{10.31}$$

where $n(\vec{r})$ is the density operator given by Eq. (5.11). For a liquid with a constant particle number density ρ,

$$\langle \chi(\vec{b}) \rangle = - \frac{2\pi a}{k_i} \rho(\vec{b}) \ell(\vec{b}) \tag{10.32}$$

where $\ell(\vec{b})$ is the length of the target parallel to $\vec{k}_i$ and going through the point $\vec{b}$, and

$$\rho(\vec{b}) = \begin{cases} \rho & \text{if } \vec{b} \text{ lies inside the target} \\ 0 & \text{otherwise} \end{cases} \tag{10.33}$$

The fluctuation term, $\langle \chi(\vec{b})\chi(\vec{b}') \rangle - \langle \chi(\vec{b}) \rangle \langle \chi(\vec{b}') \rangle$, becomes

$$\langle \chi(\vec{b})\chi(\vec{b}') \rangle - \langle \chi(\vec{b}) \rangle \langle \chi(\vec{b}') \rangle = \left(\frac{2\pi a}{k_i} \right)^2 \int dz_1 \int dz_2 \, \Gamma(\vec{b},z_1;\vec{b}',z_2) \tag{10.34}$$

where $\Gamma(\vec{r}_1,\vec{r}_2)$ is the density-density correlation function given by Eq. (5.16). Assuming a translationally invariant target (see Chapter VI),

$$\langle \chi(\vec{b})\chi(\vec{b}') \rangle - \langle \chi(\vec{b}) \rangle \langle \chi(\vec{b}') \rangle = \left[\frac{2\pi a}{k_i} \rho(\vec{b}) \right]^2 \ell(\vec{b}) \int dz \, \Gamma(\vec{b}-\vec{b}',z)/\rho^2 \tag{10.35}$$

where $z = z_1 - z_2$.

The inelastic DCS, Eq. (10.24), from Eqs. (10.32) and (10.35), is now

$$\left(\frac{d\sigma}{d\Omega}\right)_i = \left(\frac{k_i}{2\pi}\right)^2 \int_V d^2b \int_V d^2b' \, e^{i\vec{k}\cdot(\vec{b}-\vec{b}')} \, e^{i\frac{2\pi a\rho}{k_i}[\ell(\vec{b})-\ell(\vec{b}')]}$$

$$\times e^{-\frac{1}{2}\left(\frac{2\pi a}{k_i}\right)^2 [\ell(\vec{b})+\ell(\vec{b}')]\int dz\, \Gamma(z)}$$

$$\times\left\{e^{\left(\frac{2\pi a}{k_i}\right)^2 \ell(\vec{b})\int dz\, \Gamma(\vec{b}-\vec{b}';z)} - 1\right\} \tag{10.36}$$

where the limits of the b and b′ integrations now go over the target.

At this point, it is convenient to define the Fourier transform, $S(\vec{k}_1)$, of $\Gamma(\vec{r})/\rho$,

$$\Gamma(\vec{r})/\rho = (2\pi)^{-3} \int d^3k_1 \, S(\vec{k}_1) \, e^{-i\vec{k}_1\cdot\vec{r}} \tag{10.37}$$

Thus,

$$\int dz\, \Gamma(z) = \frac{\rho}{(2\pi)^2} \int d^2\lambda \, S(\vec{\lambda}) \tag{10.38}$$

and

$$\int dz\ \Gamma(\vec{b}-\vec{b}',z) = \frac{\rho}{(2\pi)^2} \int d^2\lambda\ S(\vec{\lambda})\ e^{-i\vec{\lambda}\cdot(\vec{b}-\vec{b}')} \tag{10.39}$$

where $\vec{\lambda}$ is a two dimensional wave vector perpendicular to the z direction. The inelastic DCS, Eq. (10.36), is now given by

$$\left(\frac{d\sigma}{d\Omega}\right)_i = \left(\frac{k_i}{2\pi}\right)^2 \int_V d^2b \int_V d^2b'\ e^{i\vec{k}\cdot(\vec{b}-\vec{b}')}\ e^{i\frac{2\pi a\rho}{k_i}[\ell(\vec{b})-\ell(\vec{b}')]}$$

$$\times e^{-\frac{1}{2}\frac{a^2\rho}{k_i^2}[\ell(\vec{b})+\ell(\vec{b}')]\int d^2\lambda\ S(\vec{\lambda})}$$

$$\times \left\{ e^{\frac{a^2\rho}{k_i^2}\ell(\vec{b})\int d^2\lambda\ S(\lambda)\ e^{-i\vec{\lambda}\cdot(\vec{b}-\vec{b}')}} - 1 \right\} \tag{10.40}$$

Neglecting the self-correlation term, from Eqs. (5.23) and (5.52), the correlation function, $\Gamma(r)/\rho^2$, for critical scattering, as determined by Ornstein and Zernike, is

$$\Gamma(r)/\rho^2 = \frac{1}{4\pi\rho r_1^2}\frac{e^{-\kappa_1 r}}{r} \tag{10.41}$$

Thus, from Eqs. (10.37),

$$S(\lambda) = \frac{1}{r_1^2(\lambda^2+\kappa_1^2)} \tag{10.42}$$

A formal difficulty now arises, due to the explicit form of the OZ correlation function, in that the two dimensional integral on the right-hand-side of Eq. (10.38) is divergent if taken over all values of $\vec{\lambda}$. However, recalling the meaning of $\vec{\lambda}$ as the wave vector transfer of the neutron, a natural cut-off for $\vec{\lambda}$ for quasielastic scattering is $\lambda = 2k_i$. With this cut-off for $\vec{\lambda}$, this integral becomes

$$\int d^2\lambda \; S(\vec{\lambda}) = \frac{\pi}{r_1^2} \ell n\left(1 + \frac{4k_i^2}{\kappa_1^2}\right) = \frac{k_i^2}{a^2} \sigma_B \tag{10.43}$$

where σ_B is the inelastic total cross-section per target atom as given by the first Born approximation, i.e., from Eq. (5.53),

$$\sigma_B = \int d\Omega \, \frac{1}{N} \left(\frac{d\sigma}{d\Omega}\right)_i^{(1)} = \frac{\pi a^2}{k_i^2 r_1^2} \; \ell n\left(1 + \frac{4k_i^2}{\kappa_1^2}\right) \tag{10.44}$$

In considering the inelastic DCS, Eq. (10.40), for simplicity the target is assumed to be a slab, for which $\ell(\vec{b}) = \ell$, the length of the slab parallel to $\vec{k}_i$. Notice, that with this assumption, the index of refraction term in Eq. (10.40) becomes unity. This is a reflection of the physical fact that, since all neutrons enter the target at the same z-component and then are assumed to continue moving in a straight line parallel to the z-axis, any two neutrons at some point in the target will have suffered the same target-medium-induced phase change. Thus, this phase change cannot give rise to interference effects and can have no effect on the scattering. The inelastic DCS, Eq. (10.40), for a slab target is, from Eq. (10.43),

$$\left(\frac{d\sigma}{d\Omega}\right)_i = \left(\frac{k_i}{2\pi}\right)^2 e^{-\rho\sigma_B \ell} \int_V d^2b \int_V d^2b' \, e^{i\vec{k}\cdot(\vec{b}-\vec{b}')}$$

$$\times \left\{ e^{\frac{a^2\rho\ell}{k_i^2}\int d^2\lambda \, S(\vec{\lambda}) \, e^{-i\vec{\lambda}\cdot(\vec{b}-\vec{b}')}} - 1 \right\} \qquad (10.45)$$

This is an exact result for critical nuclear scattering within the Glauber approximation and assuming slab geometry. Neutron extinction is now embodied in the simple term exp $(-\rho\sigma_B\ell)$, and all orders of scattering are represented by the terms in brackets. Expanding the exponential in the brackets leads to a series expansion of the inelastic DCS, whose terms can be identified with single and multiple scattering. Thus, the single scattering inelastic DCS is

$$\left(\frac{d\sigma}{d\Omega}\right)_i^{(1)} = Na^2 \, e^{-\rho\sigma_B\ell} \, S(k) = \frac{Na^2 \, e^{-\rho\sigma_B\ell}}{r_1^2[k^2 + \kappa_1^2]} \qquad (10.46)$$

which, neglecting the extinction factor, exp $(-\rho\sigma_B\ell)$, is the first Born or single scattering result, Eq. (5.53). The double scattering inelastic DCS is[35]

$$\left(\frac{d\sigma}{d\Omega}\right)_i^{(2)} = \left(\frac{a^2\rho}{k_i}\right)^2 V \left(\frac{\ell}{2}\right) e^{-\rho\sigma_B\ell} \int d^2\lambda \, S(\vec{\lambda}) S(\vec{k} - \vec{\lambda})$$

$$= \frac{Na^4\rho}{r_1^4}\left(\frac{\ell}{2}\right) e^{-\rho\sigma_B\ell} \, G(k_i, \theta_S, \kappa_1) \qquad (10.47)$$

where the function G is given by Eqs. (6.38) - (6.40). Thus, to second order, the inelastic DCS is

$$\left(\frac{d\sigma}{d\Omega}\right)_i = \left(\frac{d\sigma}{d\Omega}\right)_i^{(1)} + \left(\frac{d\sigma}{d\Omega}\right)_i^{(2)} = \left(\frac{d\sigma}{d\Omega}\right)_i^{(1)} [1 + \beta H(x)] \tag{10.48}$$

where β is given by

$$\beta = \frac{\pi \left(\frac{\ell}{2}\right) \rho a^2}{k_i^2 r_1^2} \tag{10.49}$$

and H(x) is given by Eq. (6.44). Neglecting the extinction factor, these results agree exactly with the previous results obtained for small-angle critical nuclear double scattering obtained from first principles in Chapter VI. Neglecting the extinction and refraction terms from the outset and calculating the double scattering term for arbitrary target geometry leads to the same results, but where now, the factor $\ell/2$ in β, Eq. (10.49), is replaced by

$$\frac{\ell}{2} \rightarrow \langle \ell \rangle = \frac{1}{2} V^{-1} \int_V d^2b \; \ell^2(\vec{b}) \tag{10.50}$$

which is equivalent[36] to the $\langle \ell \rangle$ given by Eq. (6.36) in Chapter VI.

Critical Magnetic Scattering

The formalism of the first section of this chapter is now applied to the case of critical magnetic scattering of unpolarized neutrons from

a ferromagnet slightly above its Curie temperature. For an unpolarized neutron beam, the inelastic DCS from Eq. (10.16) is, summing over final neutron spin states and averaging over initial neutron spin states,

$$\left(\frac{d\sigma}{d\Omega}\right)_i = \left(\frac{k_i}{2\pi}\right)^2 \int d^2b \int d^2b' \; e^{i\vec{k}\cdot(\vec{b}-\vec{b}')}$$

$$\times \sum_{\sigma_i} \frac{1}{2}\langle\sigma_i| \langle e^{i\chi(\vec{b})} \; e^{-i\chi(\vec{b}')}\rangle - \langle e^{i\chi(\vec{b})}\rangle\langle e^{-i\chi(\vec{b}')}\rangle|\sigma_i\rangle \qquad (10.51)$$

where $|\sigma_i\rangle$ is the initial neutron spin state.

Considering targets where the orbital contribution to the target electron's magnetic moment is negligible or can simply be taken account of by adjusting the electronic spin quantum number, the interaction, ϕ, which determines the phase shift $\chi(\vec{b})$, Eq. (10.12), is given by Eq. (7.13). Expressing ϕ in terms of its Fourier transform, $\hat{\phi}$, i.e.,

$$\phi(\vec{r}) = (2\pi)^{-3} \int d^3k_\perp \; e^{i\vec{k}_\perp\cdot\vec{r}} \; \hat{\phi}(\vec{k}_\perp) \qquad (10.52)$$

where $\hat{\phi}$ is given by Eqs. (7.16), (7.31) and (7.32), the phase factor $\chi(\vec{b})$ from Eq. (10.12) becomes

$$\chi(\vec{b}) = \frac{ge^2}{\pi m_e c^2 k_i} \sum_j \int d^2\lambda \; e^{i\vec{\lambda}\cdot(\vec{b}-\vec{r}_j)} \; \vec{s}_N\cdot\vec{p}_j(\hat{\lambda}) \qquad (10.53)$$

where $\vec{p}_j(\hat{\lambda})$ is given by Eq. (7.35),

$$\vec{p}_j(\hat{\lambda}) = \vec{s}_j - \hat{\lambda}(\vec{s}_j \cdot \hat{\lambda}) \tag{10.54}$$

Now, the same assumptions presented in Chapter VII are made, i.e., that the crystal is made up of identical atoms (with one atom per unit cell) whose nuclear thermal motions can be neglected, that electronic excitations can be neglected and that the electronic spin states are adequately described by the Heisenberg model, which ascribes to each atom an effective spin operator, $\vec{S}$, of fixed length. Expanding Eq. (10.51) in a cumulant expansion, the first cumulant vanishes for a crystal slightly above its Curie temperature, since $\langle \vec{S} \rangle = 0$, and therefore, the second cumulant becomes virtually exact for small-angle scattering for the same reasons given for the nuclear case. Thus,

$$\left(\frac{d\sigma}{d\Omega}\right)_i = \left(\frac{k_i}{2\pi}\right)^2 \int_V d^2b \int_V d^2b' \; e^{i\vec{k}\cdot(\vec{b}-\vec{b}')}$$

$$\times \sum_{\sigma_i} \frac{1}{2} \langle \sigma_i | e^{-\frac{1}{2}\langle \chi^2(\vec{b}) \rangle} e^{-\frac{1}{2}\langle \chi^2(\vec{b}') \rangle}$$

$$\times \left(e^{\langle \chi(\vec{b})\chi(\vec{b}') \rangle} - 1 \right) | \sigma_i \rangle \tag{10.55}$$

where

$$\langle \chi(\vec{b})\chi(\vec{b}') \rangle = \left(\frac{2ge^2\rho}{m_e c^2 k_i}\right)^2 \ell(\vec{b}) \int d^2\lambda \; |F(\lambda)|^2 \; e^{-i\vec{\lambda}\cdot(\vec{b}-\vec{b}')}$$

$$\times \int d^3R \; e^{i\vec{\lambda}\cdot\vec{R}} \langle \vec{s}_N \cdot \vec{P}_0(\hat{\lambda}) \vec{s}_N \cdot \vec{P}_{\vec{R}}(-\hat{\lambda}) \rangle \; , \tag{10.56}$$

$$\vec{P}_{\vec{R}}(\hat{\lambda}) = \vec{S}_{\vec{R}} - \hat{\lambda}(\vec{S}_{\vec{R}}\cdot\hat{\lambda}) \tag{10.57}$$

is the effective Heisenberg spin in the direction perpendicular to $\hat{\lambda}$ of an atom located at $\vec{R}$, Eq. (7.47), and $F(\lambda)$ is the magnetic atomic form factor, Eq. (7.52). In obtaining Eq. (10.56), the spin system was assumed to be translationally invariant and the sums over the location of the Heisenberg spin operators were replaced with integrals, retaining only the $\vec{\tau} = 0$ reciprocal lattice vector. The details of this calculation are left to the interested reader since it closely parallels the calculation in Chapter VII.

Now making use of the invariance of the Heisenberg model Hamiltonian to simultaneous rotations of the target spins, Eq. (7.58), the target spin average appearing in Eq. (10.56) can be rewritten as,

$$\langle \vec{s}_N\cdot\vec{P}_0(\hat{\lambda})\vec{s}_N\cdot\vec{P}_{\vec{R}}(-\hat{\lambda})\rangle = \frac{1}{3}\vec{p}_N(\hat{\lambda})\cdot\vec{p}_N(-\hat{\lambda})\langle\vec{S}_0\cdot\vec{S}_{\vec{R}}\rangle = \frac{1}{6}\langle\vec{S}_0\cdot\vec{S}_{\vec{R}}\rangle \tag{10.58}$$

where

$$\vec{p}_N(\hat{\lambda}) = \vec{s}_N - \hat{\lambda}(\vec{s}_N\cdot\hat{\lambda}) \tag{10.59}$$

represents the projection of the neutron spin perpendicular to $\vec{\lambda}$. Since $\vec{p}_N\cdot\vec{p}_N = \frac{1}{2}$, a c-number, the inelastic DCS, Eq. (10.51), is independent of the initial neutron spin state and the average over initial neutron spin states appearing in the DCS, Eq. (10.55), is unity.

Defining the Fourier transform $S(\vec{k}_1)$ of $\langle \vec{S}_0 \cdot \vec{S}_{\vec{R}} \rangle / \rho$,

$$\langle \vec{S}_0 \cdot \vec{S}_{\vec{R}} \rangle / \rho = (2\pi)^{-3} \int d^3k_1 \; S(\vec{k}_1) \; e^{-i\vec{k}_1 \cdot \vec{R}} , \tag{10.60}$$

gives

$$\int d^3R \; e^{i\vec{\lambda} \cdot \vec{R}} \; \langle \vec{S}_0 \cdot \vec{S}_{\vec{R}} \rangle = \rho S(\vec{\lambda}) \tag{10.61}$$

and

$$\langle \chi(\vec{b}) \chi(\vec{b}') \rangle = \frac{2}{3} \left(\frac{ge^2}{m_e c^2 k_i} \right)^2 \rho \ell(\vec{b}) \int d^2\lambda \, |F(\lambda)|^2 \; e^{-i\vec{\lambda} \cdot (\vec{b} - \vec{b}')} \; S(\vec{\lambda}) \tag{10.62}$$

Near the critical point, $\langle \vec{S}_0 \cdot \vec{S}_{\vec{R}} \rangle$ as given by Ornstein and Zernike for large R, Eqs. (7.75) and (7.84), is

$$\langle S_0 \cdot S_{\vec{R}} \rangle = \frac{S(S+1)}{4\pi\rho r_1^2} \; \frac{e^{-\kappa_1 R}}{R} \tag{10.63}$$

and

$$S(\vec{\lambda}) = \frac{S(S+1)}{r_1^2} \; \frac{1}{\lambda^2 + \kappa_1^2} \tag{10.64}$$

Since $S(\vec{\lambda})$ is appreciable in the critical region only for small values of λ for which $F(\lambda) \cong 1$, the magnetic form factor in Eq. (10.62) is replaced by unity. Thus,

$$\langle \chi(\vec{b})\chi(\vec{b}') \rangle = \frac{2}{3} S(S+1) \left(\frac{ge^2}{m_e c^2}\right)^2 \frac{\rho \ell(\vec{b})}{k_i^2} \int d^2\lambda \; [r_1^2(\lambda^2 + \kappa_1^2)]^{-1}$$

$$\times \; e^{-i\vec{\lambda}\cdot(\vec{b}-\vec{b}')} \qquad (10.65)$$

Comparing Eq. (10.65) with the treatment for critical nuclear scattering from liquids presented in the second section of this chapter, it becomes apparent that, as before [see Eq. (8.35)], the critical magnetic scattering can be obtained from the nuclear scattering upon replacement of the nuclear scattering length with the corresponding magnetic scattering length

$$a^2 \rightarrow \frac{2}{3} S(S+1) \left(\frac{ge^2}{m_e c^2}\right)^2 . \qquad (10.66)$$

This replacement made in Eqs. (10.46) - (10.49) gives the single and double magnetic inelastic DCS. In particular, the double scattering parameter β, Eq. (10.49), becomes, for magnetic scattering,

$$\beta = \frac{2\pi\left(\frac{1}{2}\ell\right)\rho S(S+1)}{3k_i^2 r_1^2} \left(\frac{ge^2}{m_e c^2}\right)^2 \qquad (10.67)$$

which agrees exactly with the results obtained in Chapter VIII, see Eq. (8.38).

Critical Multiple Scattering in the Asymptotic Limit $\kappa_1 \to 0$

Up to now, this entire treatment has dealt mainly with the expansion of the scattering cross section in a perturbative series and considering in detail only terms through double scattering. However, with the Glauber approximation, it is now possible to obtain the cross section for critical scattering in a closed form, in the asymptotic limit $\kappa_1 \to 0$, that is useful for analysis of experimental data. In the first section of this chapter, a plausibility argument was given for the applicability of the GA to critical scattering of thermal-neutrons. Subsequently, it was shown that indeed the GA did produce exactly the same results, through double scattering, as was previously obtained from first principles. Therefore, in what follows, it is implicitly assumed that the GA is applicable to critical scattering for all orders of scattering. While this assertion has by no means been proved, it does seem very reasonable.

Assuming slab geometry, the inelastic DCS for critical nuclear or magnetic scattering is

$$\left(\frac{d\sigma}{d\Omega}\right)_i = A\left(\frac{k_i}{2\pi}\right)^2 e^{-\rho\sigma_B \ell} \int d^2b'' \, e^{i\vec{k}\cdot\vec{b}''} \times \left\{ e^{\frac{2\beta}{\pi}\int d^2\lambda \, (\lambda^2 + \kappa_1^2)^{-1} e^{-\vec{\lambda}\cdot\vec{b}''}} - 1 \right\} \qquad (10.68)$$

where A is the cross-sectional area of the target and β is given by Eq. (10.49) for nuclear scattering and by Eq. (10.67) for magnetic scat-

tering. The integral over λ appearing in Eq. (10.68) can be expressed in terms of the modified Bessel function K_0,[37]

$$\frac{2\beta}{\pi}\int d^2\lambda\ (\lambda^2 + \kappa_1^2)^{-1}\ e^{-i\vec{\lambda}\cdot\vec{b}''} = 4\beta K_0(\kappa_1 b'') \tag{10.69}$$

The inelastic DCS, Eq. (10.68), can then be written

$$\left(\frac{d\sigma}{d\Omega}\right)_i = \frac{Ak_i^2}{2\pi}\, e^{-\rho\sigma_B \ell} \int_0^\infty db''b''\ J_0(kb'')\left[e^{4\beta K_0(\kappa_1 b'')} - 1\right] \tag{10.70}$$

where J_0 is the zeroth-order Bessel function of the first kind.

As the critical point is approached, $\kappa_1 \to 0$ and the integral in Eq. (10.70) is easily evaluated in this limit. For $\kappa_1 \to 0$ and $k \neq 0$, the asymptotic expansion for $K_0(\kappa_1 b'')$ in this limit is used and the dependence of the inelastic DCS on wave vector transfer k is given by[38]

$$\left(\frac{d\sigma}{d\Omega}\right)_i \xrightarrow[\kappa_1 \to 0]{} \frac{Ak_i^2}{\pi\kappa_1^2}\, e^{-\rho\sigma_B \ell}\, \frac{\Gamma(1-2\beta)}{\Gamma(2\beta)} \left(\frac{1}{k\kappa_1^{-1}}\right)^{2-4\beta} \qquad \left(\beta < \frac{1}{2}\right) \tag{10.71}$$

For the typically small experimental values of β, the ratio of Γ functions in Eq. (10.71) can be replaced by 2β, yielding

$$\left(\frac{d\sigma}{d\Omega}\right)_i \simeq \frac{2\beta Ak_i^2}{\pi\kappa_1^2}\, e^{-\rho\sigma_B \ell} \left(\frac{1}{k\kappa_1^{-1}}\right)^{2-4\beta} \tag{10.72}$$

Compared with the single scattering cross section for $\kappa_1 \to 0$, from Eq. (10.46),

$$\left(\frac{d\sigma}{d\Omega}\right)_i^{(1)} \cong \frac{2\beta A k_i^{\,2}}{\pi \kappa_1^{\,2}} e^{-\rho\sigma_B \ell} \left(\frac{1}{k\kappa_1^{\,-1}}\right)^2 \tag{10.73}$$

it appears that multiple scattering effects can be accounted for by the simple replacement of the exponent on the $1/k\kappa_1^{\,-1}$ factor in the single scattering inelastic DCS of

$$2 \to 2 - 4\beta \tag{10.74}$$

Equation (10.72) has the same form as Fisher's modified form for the inelastic DCS, in the limit $\kappa_1 \to 0$, [see Eq. (9.10)] and obviously gives a multiple scattering induced η of

$$\eta = 4\beta \tag{10.75}$$

This result is in agreement with the results obtained in Chapter IX [see Eq. (9.22)] in the limit $\kappa_1 \to 0$.

CHAPTER XI

CONCLUSIONS AND RECOMMENDATIONS

In this work, a general quantum mechanical multiple scattering theory has been formulated for thermal-neutron scattering from macroscopic materials and applied to single and double scattering. Thermal-neutron critical scattering from liquids and ferromagnets slightly above their critical points was treated in detail, and resulted in a critical double scattering parameter, β, from which the magnitude of double scattering effects can be determined. For typical experimental conditions, critical magnetic double scattering from ferromagnets was found to produce small, but non-negligible deviations in the single scattering Lorentzian line shape of scattered neutrons. In this regard, it competes with similar line shape deviations predicted by Fisher's modification to the correlation function from the OZ form used in the single scattering cross section.

In addition, Glauber's high energy approximation was shown to provide a simple description for thermal-neutron critical multiple scattering which produced results in agreement with those previously obtained from first principles. When sufficiently close to the critical point, the GA was shown to provide a closed-form expression for the cross section, containing all orders of scattering, which has the same form as the single scattering cross section with a modified exponent for the wave vector transfer. Compared to Fisher's modification of the

single scattering cross section, this result gives an apparent, multiple scattering induced η of magnitude 4β.

The general scattering formalism presented in Chapters II - IV will accommodate other types of scattering, such as light and x-ray scattering. Since critical x-ray scattering, like thermal-neutron scattering, is predominately in the forward direction, the critical scattering formalism presented here is also applicable to x-rays. However, critical light scattering (in the optical region) is not restricted to the forward direction; therefore, determination of the target-size parameter $\langle \ell \rangle$ is more complicated. In addition, extinction effects due to the absorption of photons can be appreciable and must be taken into account.

Critical magnetic scattering from anti-ferromagnets is pronounced in non-forward directions around magnetic Bragg peak positions. Therefore, double scattering appears to make a negligible contribution. In addition, all small-angle approximations made in the double scattering theory, e.g., in the determination of $\langle \ell \rangle$ and setting the magnetic form factors equal to unity, are invalid for scattering from anti-ferromagnets. The magnitude of the multiple scattering also depends on such experimental details as, for example, the orientation of the anti-ferromagnetic crystal with respect to the wave vector transfer. Therefore, the application of the theory presented here to critical magnetic scattering from anti-ferromagnets appears to be uninteresting unless one is analyzing a particular scattering experiment.

Although only multiple scattering using unpolarized neutron

beams was considered here, the theory can be extended to include scattering of polarized neutrons which will reveal more detailed information about the target structure. Also, small-angle thermal-neutron multiple scattering from low-temperature spin waves might prove to be a good candidate for further investigations.

Only quasielastic scattering was considered in detail in this treatment. Thus, applications of the most powerful aspect of the general multiple scattering theory presented in Chapters II - IV was completely ignored, i.e., the ability to treat inelastic ($\omega \neq 0$) multiple scattering. Within the QA, a T-matrix expansion would have produced, perhaps even in a simpler, more efficient way, the same results for critical scattering obtained here. Therefore, in order that the full potential of the theory presented here be realized, inelastic scattering effects, for both critical scattering and scattering under general conditions, should be considered.

Portions of this work are contained in two articles published in Physical Review B by H. A. Gersch and this author.[39,40]

APPENDIX I

THE TIME ORDERING OPERATOR

If the operators H_o and ϕ in the exponential $\exp\left[-i(H_o + \phi)\times (t - t_o)/\hbar\right]$ do not commute, then the exponential cannot be written as a simple product of exponentials. However, it is often convenient, and necessary, to write this exponential as a product of terms, each term containing only one of the operators. This is possible by utilizing time ordering operators.

In this appendix it is shown that

$$e^{-i(H_o+\phi)(t-t_o)/\hbar} = e^{-iH_o t/\hbar}\left[T_+ e^{-\frac{i}{\hbar}\int_{t_o}^{t} dt_1\, \phi(t_1)}\right] e^{iH_o t_o/\hbar} \tag{I.1}$$

To show this, a function $\Lambda(t)$ is defined such that

$$e^{-i(H_o+\phi)(t-t_o)/\hbar} = e^{-iH_o(t-t_o)/\hbar}\,\Lambda(t) \tag{I.2}$$

The problem now becomes one of determining the functional form of $\Lambda(t)$. Taking the time derivative of Eq. (I.2) and multiplying the resulting equation by $\exp\left[iH_o(t - t_o)/\hbar\right]$ yields

$$- \frac{i}{\hbar} H_o \, e^{iH_o(t-t_o)/\hbar} \, e^{-i(H_o+\phi)(t-t_o)/\hbar} - \frac{i}{\hbar} e^{iH_o(t-t_o)/\hbar}$$

$$\times \phi \, e^{-i(H_o+\phi)(t-t_o)/\hbar} = - \frac{i}{\hbar} H_o \Lambda(t) + \frac{d\Lambda(t)}{dt} \tag{I.3}$$

From Eq. (I.2),

$$\Lambda(t) = e^{iH_o(t-t_o)/\hbar} \, e^{-i(H_o+\phi)(t-t_o)/\hbar} \tag{I.4}$$

Therefore, Eq. (I.3) becomes

$$\frac{d\Lambda(t)}{dt} = - \frac{i}{\hbar} e^{iH_o(t-t_o)/\hbar} \, \phi \, e^{-i(H_o+\phi)(t-t_o)/\hbar} \tag{I.5}$$

Inserting the identity operator

$$I = e^{-iH_o(t-t_o)\hbar} \, e^{iH_o(t-t_o)/\hbar} \tag{I.6}$$

into Eq. (I.5) and using Eq. (I.4), Eq. (I.5) becomes

$$\frac{d\Lambda(t)}{dt} = - \frac{i}{\hbar} \phi(t-t_o)\Lambda(t) \tag{I.7}$$

where

$$\phi(t-t_o) = e^{iH_o(t-t_o)} \phi\, e^{-iH_o(t-t_o)} \tag{I.8}$$

is in the interaction picture. Integrating Eq. (I.7), and using the initial condition $\Lambda(t_o) = 1$, from Eq. (I.4), yields

$$\Lambda(t) = 1 - \frac{i}{\hbar} \int_{t_o}^{t} dt_1 \phi(t_1-t_o)\Lambda(t_1) \tag{I.9}$$

or, after successive iterations,

$$\Lambda(t) = 1 - \frac{i}{\hbar} \int_{t_o}^{t} dt_1 \phi(t_1-t_o) + \left(-\frac{i}{\hbar}\right)^2 \int_{t_o}^{t} dt_1 \int_{t_o}^{t_1} dt_2\, \phi(t_1-t_o)\phi(t_2-t_o)$$

$$+ \ldots \tag{I.10}$$

Using Eq. (I.8), Eq. (I.10) may be rewritten as

$$\Lambda(t) = e^{-iH_o t_o/\hbar} \left\{ 1 - \frac{i}{\hbar} \int_{t_o}^{t} dt_1 \phi(t_1) \right.$$

$$\left. + \left(-\frac{i}{\hbar}\right)^2 \int_{t_o}^{t} dt_1 \int_{t_o}^{t_1} dt_2 \phi(t_1)\phi(t_2) + \ldots \right\} e^{iH_o t_o/\hbar} \tag{I.11}$$

The time ordering operator, T_+, is defined such that

$$T_+\left[\phi(t_1)\phi(t_2)\right] = \begin{cases} \phi(t_1)\phi(t_2) & t_1 \geq t_2 \\ \phi(t_2)\phi(t_1) & t_2 \geq t_1 \end{cases} \tag{I.12}$$

Notice that,

$$T_+\left[\int_{t_o}^{t} dt_1\ \phi(t_1)\right]^2 = T_+\left[\int_{t_o}^{t} dt_1 \int_{t_o}^{t} dt_2\ \phi(t_1)\phi(t_2)\right]$$

$$= T_+\left[\int_{t_o}^{t} dt_1 \int_{t_o}^{t_1} dt_2\ \phi(t_1)\phi(t_2) + \int_{t_o}^{t_2} dt_1 \int_{t_o}^{t} dt_2\ \phi(t_1)\phi(t_2)\right] \tag{I.13}$$

where the mathematical identity

$$\int_o^a dx \int_o^b dy\ f(x)g(x) = \int_o^a dx \int_o^x dy\ f(x)g(y)$$

$$+ \int_o^y dx \int_o^b dy\ f(x)g(y) \tag{I.14}$$

has been utilized. Therefore, performing the time ordering and making a change of integration variables,

$$T_+\left[\int_{t_o}^{t} dt_1\ \phi(t_1)\right]^2 = 2\int_{t_o}^{t} dt_1 \int_{t_o}^{t_1} dt_2\ \phi(t_1)\phi(t_2) \tag{I.15}$$

Similarly, it can be shown that

$$T_+\left[\int_{t_o}^{t} dt_1\ \phi(t_1)\right]^n =$$

$$= n!\int_{t_o}^{t} dt_1 \int_{t_o}^{t_1} dt_2 \ \ldots \int_{t_o}^{t_{n-1}} dt_n\ \phi(t_1)\phi(t_2)\ \ldots\ \phi(t_n) \qquad \text{(I.16)}$$

Therefore, Eq. (I.11) can be written as

$$\Lambda(t) = e^{-iH_o t_o/\hbar}\left[T_+ \sum_{n=0}^{\infty} \frac{1}{n!}\left(-\frac{i}{\hbar}\int_{t_o}^{t} dt_1\ \phi(t_1)\right)^n\right] e^{iH_o t_o/\hbar} \qquad \text{(I.17)}$$

or, more compactly and formally as

$$\Lambda(t) = e^{-iH_o t_o/\hbar}\left[T_+\, e^{-\frac{i}{\hbar}\int_{t_o}^{t} dt_1\ \phi(t_1)}\right] e^{iH_o t_o/\hbar} \qquad \text{(I.18)}$$

Substituting this expression for $\Lambda(t)$ back into Eq. (I.2) yields

$$e^{-i(H_o+\phi)(t-t_o)/\hbar} = e^{-iH_o t/\hbar}\left[T_+\, e^{-\frac{i}{\hbar}\int_{t_o}^{t} dt_1\ \phi(t_1)}\right] e^{iH_o t_o/\hbar} \qquad \text{(I.19)}$$

which is Eq. (I.1).

Making the identification $H_o = H_N + H_T$ in Eq. (I.19) gives Eq. (3.14) in the text, and Eq. (3.15) in the text can be obtained from the same analysis presented in this appendix.

APPENDIX II

CORRELATIONS OF FLUCTUATIONS OF AN EXTENSIVE VARIABLE

Consider a system with fixed volume V divided up into M identical cells. Consider one arbitrary extensive variable, y, (other than V) of the system, holding all other variables fixed, and let y_k denote the value of y in the k^{th} cell. A statistical state of the system is given by specifying the value of y in all cells, i.e., by the set of numbers $\{y_k\}$. Assume that the free energy, F, of the system is given by the quadratic form

$$F = \frac{1}{2} C' \sum_{k,\ell=1}^{M} a_{k\ell} z_k z_\ell \tag{II.1}$$

where C' is a constant and the constant $a_{k\ell}$ is a measure of the interaction between the k^{th} cell and the ℓ^{th} cell, and is symmetric, i.e., $a_{k\ell} = a_{\ell k}$. The variable z_k is the fluctuation of y_k about its mean value y, i.e.,

$$z_k = y_k - y \tag{II.2}$$

The probability, P, of finding the system in state $\{y_k\}$ is given by

$$P = C\, e^{-\frac{\theta}{2} \sum_{k,\ell} a_{k\ell} z_k z_\ell} \tag{II.3}$$

where C is a constant, and

$$\theta = (k_B T)^{-1} \tag{II.4}$$

Considering $a_{k\ell}$ to be the elements of a matrix A, i.e.,

$$A = \begin{bmatrix} a_{11} & a_{12} & \cdots & a_{1M} \\ a_{21} & & & \\ \vdots & & \ddots & \\ a_{M1} & & & a_{MM} \end{bmatrix} \tag{II.5}$$

then, in matrix notation,

$$\sum_{k,\ell} a_{k\ell} z_k z_\ell = \vec{z}^T A \vec{z} \tag{II.6}$$

where $\vec{z}$ is the column matrix

$$\vec{z} = \begin{bmatrix} z_1 \\ z_2 \\ \vdots \\ z_M \end{bmatrix} \tag{II.7}$$

and $\vec{z}^T$ is the transpose of $\vec{z}$.

In order to express Eq. (II.6) in terms of normal coordinates, $\{\alpha_k\}$, the following eigenvalue equation is solved for its eigenvectors, $\{\vec{b}_m\}$, and eigenvalues, $\{\lambda_m\}$

$$A\vec{b}_m = \lambda_m \vec{b}_m \tag{II.8}$$

where

$$\vec{b}_m = \begin{bmatrix} b_{1m} \\ b_{2m} \\ \cdot \\ \cdot \\ \cdot \\ b_{Mm} \end{bmatrix} \tag{II.9}$$

Since A is symmetric, i.e., $A = A^T$, its eigenvectors are orthogonal, i.e.,

$$\vec{b}_k^T \vec{b}_\ell = c\ \delta_{k,\ell} \tag{II.10}$$

where c is a constant. Equation (II.8) does not uniquely determine the eigenvectors; therefore, a normalization condition can be imposed. Requiring $c = 1$,

$$\vec{b}_k^T \vec{b}_\ell = \delta_{k,\ell} \tag{II.11}$$

The normal coordinates, $\{\alpha_k\}$, are defined by

$$\vec{z} = B\vec{\alpha} \tag{II.12}$$

where

$$B = \begin{bmatrix} b_{11} & b_{12} & \cdots & b_{1M} \\ b_{21} & & & \\ \cdot & & \cdot & \\ \cdot & & & \cdot \\ \cdot & & & \\ b_{M1} & & & b_{MM} \end{bmatrix} \tag{II.13}$$

and

$$\vec{\alpha} = \begin{bmatrix} \alpha_1 \\ \alpha_2 \\ \cdot \\ \cdot \\ \cdot \\ \alpha_M \end{bmatrix} \tag{II.14}$$

Notice, from Eq. (II.11), that the matrix B is orthogonal, i.e., $B^T = B^{-1}$. In terms of B, the eigenvalue equation, Eq. (II.8), can be written in matrix notation as

$$AB = B\lambda \tag{II.15}$$

where

$$\lambda = \begin{bmatrix} \lambda_1 & & & 0 \\ & \lambda_2 & & \\ & & \ddots & \\ 0 & & & \lambda_M \end{bmatrix} \qquad \text{(II.16)}$$

From Eqs. (II.12), (II.15) and (II.16) and using the fact that B is orthogonal, Eq. (II.6) can be written

$$\vec{z}^T A \vec{z} = \vec{\alpha}^T B^T A B \vec{\alpha} = \vec{\alpha}^T \lambda \vec{\alpha} = \sum_k \alpha_k^2 \lambda_k \qquad \text{(II.17)}$$

Thus, in terms of normal coordinates, the probability P, Eq. (II.3), becomes

$$P(\alpha) = C\, e^{-\frac{\theta}{2} \sum_k \alpha_k^2 \lambda_k} \qquad \text{(II.18)}$$

The correlation between z_k and z_ℓ is, from Eqs. (II.12) - (II.14),

$$\langle z_k z_\ell \rangle = \sum_{m,n} b_{km} b_{\ell n} \langle \alpha_m \alpha_n \rangle \qquad \text{(II.19)}$$

and in terms of $P(\alpha)$, Eq. (II.18), the correlation between α_m and α_n is given by

$$\langle \alpha_m \alpha_n \rangle = \frac{\int_{-\infty}^{\infty} \cdots \int_{-\infty}^{\infty} d\alpha_1 \cdots d\alpha_M \, \alpha_m \alpha_n \, P(\alpha)}{\int_{-\infty}^{\infty} \cdots \int_{-\infty}^{\infty} d\alpha_1 \cdots d\alpha_M \, P(\alpha)}$$

$$= \frac{\int_{-\infty}^{\infty} d\alpha_m \int_{-\infty}^{\infty} d\alpha_n \, \alpha_m \alpha_n \, e^{-\frac{\theta}{2}\alpha_m^2 \lambda_m} e^{-\frac{\theta}{2}\alpha_n^2 \lambda_n}}{\int_{-\infty}^{\infty} d\alpha_m \int_{-\infty}^{\infty} d\alpha_n \, e^{-\frac{\theta}{2}\alpha_m^2 \lambda_m} e^{-\frac{\theta}{2}\alpha_n^2 \lambda_n}}$$

$$= \delta_{m,n} (\theta \lambda_m)^{-1} \tag{II.20}$$

Therefore, Eq. (II.19) becomes

$$\langle z_k z_\ell \rangle = \theta^{-1} \sum_m \frac{b_{km} b_{\ell m}}{\lambda_m} = \theta^{-1} (B \lambda^{-1} B^T)_{k\ell} \tag{II.21}$$

or, from Eq. (II.15) and using the fact that B is orthogonal,

$$\langle z_k z_\ell \rangle = \theta^{-1} (A^{-1})_{k\ell} \tag{II.22}$$

which is Eq. (5.31) in the text.

The correlation of three z's is given by

$$\langle z_j z_k z_\ell \rangle = \sum_{m,n,p} b_{jm} b_{kn} b_{\ell p} \langle \alpha_m \alpha_n \alpha_p \rangle \tag{II.23}$$

and

$$\langle \alpha_m \alpha_n \alpha_p \rangle =$$

$$\frac{\int_{-\infty}^{\infty} d\alpha_m \int_{-\infty}^{\infty} d\alpha_n \int_{-\infty}^{\infty} d\alpha_p \; \alpha_m \alpha_n \alpha_p \; e^{-\frac{\theta}{2}\left[\alpha_m^2 \lambda_m + \alpha_n^2 \lambda_n + \alpha_p^2 \lambda_p\right]}}{\int_{-\infty}^{\infty} d\alpha_m \int_{-\infty}^{\infty} d\alpha_n \int_{-\infty}^{\infty} d\alpha_p \; e^{-\frac{\theta}{2}\left[\alpha_m^2 \lambda_m + \alpha_n^2 \lambda_n + \alpha_p^2 \lambda_p\right]}} = 0 \qquad \text{(II.24)}$$

since

$$\int_{-\infty}^{\infty} d\alpha \; \alpha^n \; e^{-\frac{\theta}{2}\alpha^2 \lambda} = 0 \qquad (n \text{ odd}) \qquad \text{(II.25)}$$

Therefore,

$$\langle z_j z_k z_\ell \rangle = 0 \qquad \text{(II.26)}$$

which is Eq. (6.7) in the text. In fact, from Eq. (II.25), it follows that the correlation of any odd number of z's is zero.

The correlation of four z's is given by

$$\langle z_i z_j z_k z_\ell \rangle = \sum_{m,n,p,q} b_{im} b_{jn} b_{kp} b_{\ell q} \langle \alpha_m \alpha_n \alpha_p \alpha_q \rangle \qquad \text{(II.27)}$$

It is easily shown, using Eqs. (II.20) and (II.25), that

$$\langle \alpha_m \alpha_n \alpha_p \alpha_q \rangle$$

$$= \begin{cases} (\theta\lambda_m)^{-1}(\theta\lambda_p)^{-1} = \langle \alpha_m \alpha_n \rangle \langle \alpha_p \alpha_q \rangle & \text{if } m = n \neq p = q \\ (\theta\lambda_m)^{-1}(\theta\lambda_n)^{-1} = \langle \alpha_m \alpha_p \rangle \langle \alpha_n \alpha_q \rangle & \text{if } m = p \neq n = q \\ (\theta\lambda_m)^{-1}(\theta\lambda_n)^{-1} = \langle \alpha_m \alpha_q \rangle \langle \alpha_n \alpha_p \rangle & \text{if } m = q \neq n = p \\ 3(\theta\lambda_m)^{-2} = \langle \alpha_m \alpha_n \rangle \langle \alpha_p \alpha_q \rangle + \langle \alpha_m \alpha_p \rangle \langle \alpha_n \alpha_q \rangle & \\ \qquad + \langle \alpha_m \alpha_q \rangle \langle \alpha_n \alpha_p \rangle & \text{if } m = n = p = q \\ 0 & \text{otherwise} \end{cases} \tag{II.28}$$

or

$$\langle \alpha_m \alpha_n \alpha_p \alpha_q \rangle = \langle \alpha_m \alpha_n \rangle \langle \alpha_p \alpha_q \rangle + \langle \alpha_m \alpha_p \rangle \langle \alpha_n \alpha_q \rangle$$

$$\langle \alpha_m \alpha_q \rangle \langle \alpha_n \alpha_p \rangle \tag{II.29}$$

Therefore, from Eq. (II.19), Eq. (II.27) becomes

$$\langle z_i z_j z_k z_\ell \rangle = \langle z_i z_j \rangle \langle z_k z_\ell \rangle + \langle z_i z_k \rangle \langle z_j z_\ell \rangle$$

$$+ \langle z_i z_\ell \rangle \langle z_j z_k \rangle \tag{II.30}$$

which is Eq. (6.8) in the text.

Similar calculations on higher order correlations of any even number of z's will show that they also decompose into sums of products of pair correlations.

APPENDIX III

EVALUATION OF THE INTEGRAL $G(k_i,\theta_S,\kappa_1)$

In this appendix, the integral

$$G(k_i,\theta_S,\kappa_1) = \int_0^{2\pi} d\phi_1 \int_0^{\pi} d\theta_1 \sin\theta_1 \left[4k_i^2 \sin^2\frac{\theta_1}{2} + \kappa_1^2\right]^{-1} \times \left[4k_i^2 \sin^2\frac{\theta_2}{2} + \kappa_1^2\right]^{-1} \qquad \text{(III.1)}$$

is evaluated in the small-angle approximation, i.e., $\sin\theta \tilde{=} \theta$. The angles θ_1 and θ_2 are the intermediate scattering angles associated with, respectively, the first and second scattering events. The angle θ_2 will depend on θ_1, ϕ_1 and the scattering angle θ_S.

Consider two coordinate systems, S and S′. The origin of the S system is located at the first scattering event with its z-axis in the direction of $\vec{k}_i$, and the origin of the S′ system is located at the second scattering event with its z′-axis in the direction of $\vec{q}$, see Figure 5a. The scattering angle, θ_S, is defined as the angle between $\vec{k}_i$ and $\vec{k}_f$, see Figure 5b. Since the distance from the target to the detector is much greater than the distance between the two scattering events, for the purpose of evaluating G the origins of S and S′ can be considered to coincide with negligible error, see Figure 5c. The angular coordinates of $\vec{k}_f$ are (θ_S,ϕ_S) as measured in S and (θ_2,ϕ_2) as

measured in S′, and the angular coordinates of $\vec{q}$ are (θ_1,ϕ_1) as measured in S.

Now, to evaluate G, the angle θ_2 must be determined in terms of the angles θ_1, ϕ_1 and θ_S. The only restraints on S and S′ so far have been that the z-axis be along $\vec{k}_i$ and that the z′-axis be along $\vec{q}$. The orientations of the x-, y-, x′- and y′-axes have not been fixed. Therefore, letting the S system be fixed, the S′ system can be generated from the S system by rotations through two Eulerian angles:[41]

(1) Rotate S counterclockwise about the z-axis through an angle $\pi/2 + \phi_1$ to obtain the x -axis, and

(2) rotate the system obtained from the above rotation about the x′-axis through an angle θ_1 to obtain the y′- and z′-axes.

Now, the coordinates (x′,y′, z′) of a point measured in S′ are related to the coordinates (x,y,z) of the same point measured in S by

$$\begin{bmatrix} x' \\ y' \\ z' \end{bmatrix} = \begin{bmatrix} -\sin\phi_1 & \cos\phi_1 & 0 \\ -\cos\phi_1\cos\theta_1 & -\sin\phi_1\cos\theta_1 & \sin\theta_1 \\ \cos\phi_1\sin\theta_1 & \sin\phi_1\sin\theta_1 & \cos\theta_1 \end{bmatrix} \begin{bmatrix} x \\ y \\ z \end{bmatrix} \qquad \text{(III.2)}$$

Writing Eq. (III.2) in terms of spherical polar coordinates, there results, for any fixed point along $\vec{k}_f$, the angular equations

$$\begin{bmatrix} \sin\theta_2 \cos\phi_2 \\ \sin\theta_2 \sin\phi_2 \\ \cos\theta_2 \end{bmatrix} =$$

$$\begin{bmatrix} -\sin\phi_1 & \cos\phi_1 & 0 \\ -\cos\phi_1 \cos\theta_1 & -\sin\phi_1 \cos\theta_1 & \sin\theta_1 \\ \cos\phi_1 \sin\theta_1 & \sin\phi_1 \sin\theta_1 & \cos\theta_1 \end{bmatrix} \begin{bmatrix} \sin\theta_S \cos\phi_S \\ \sin\theta_S \sin\phi_S \\ \cos\theta_S \end{bmatrix} \quad \text{(III.3)}$$

or,

$$\sin\theta_2 \cos\phi_2 = -\sin\phi_1 \sin\theta_S \cos\phi_S + \cos\phi_1 \sin\theta_S \sin\phi_S \quad \text{(III.4)}$$

$$\sin\theta_2 \sin\phi_2 = -\cos\phi_1 \cos\theta_1 \sin\theta_S \cos\phi_S - \sin\phi_1 \cos\theta_1 \sin\theta_S \sin\phi_S + \sin\theta_1 \cos\theta_S \quad \text{(III.5)}$$

$$\cos\theta_2 = \cos\phi_1 \sin\theta_1 \sin\theta_S \cos\phi_S + \sin\phi_1 \sin\theta_1 \sin\theta_S \sin\phi_S + \cos\theta_1 \cos\theta_S \quad \text{(III.6)}$$

These three equations are not linearly independent. Squaring Eqs. (III.4) and (III.5), then adding them together yields Eq. (III.6), which can be

written as

$$\cos\theta_2 = \sin\theta_1 \sin\theta_S \cos(\phi_1-\phi_S) + \cos\theta_1 \cos\theta_S \qquad \text{(III.7)}$$

or

$$\sin^2\frac{\theta_2}{2} = \frac{1}{2}\left[1 - \sin\theta_1 \sin\theta_S \cos(\phi_1-\phi_S) - \cos\theta_1 \cos\theta_S\right] \qquad \text{(III.8)}$$

Using Eq. (III.8), the integral G, Eq. (III.1), in the small-angle approximation, becomes

$$G(k_i,\theta_S,\kappa_1) = \int_0^{2\pi} d\phi_1 \int_0^{\infty} d\theta_1\, \theta_1$$

$$\times \left[k_i^2\theta_1^2+\kappa_1^2\right]^{-1} \left[k_i^2(\theta_1^2+\theta_S^2-2\theta_1\theta_S \cos\phi_1) + \kappa_1^2\right]^{-1} \qquad \text{(III.9)}$$

where ϕ_S has been set equal to zero, since, as can easily be shown, the integral G is independent of ϕ_S. Now, G is easily evaluated[42] and yields

$$G(k_i,\theta_S,\kappa_1) = \frac{\pi}{k_i^2\kappa_1^2 x(x+4)^{\frac{1}{2}}} \ln\left[\frac{x^3+3x+(x^2+1)(x^2+4)^{\frac{1}{2}}}{(x+4)^{\frac{1}{2}}-x}\right] \qquad \text{(III.10)}$$

where $x = k_i\theta_S\kappa_1^{-1}$, which is Eq. (6.38) in the text.

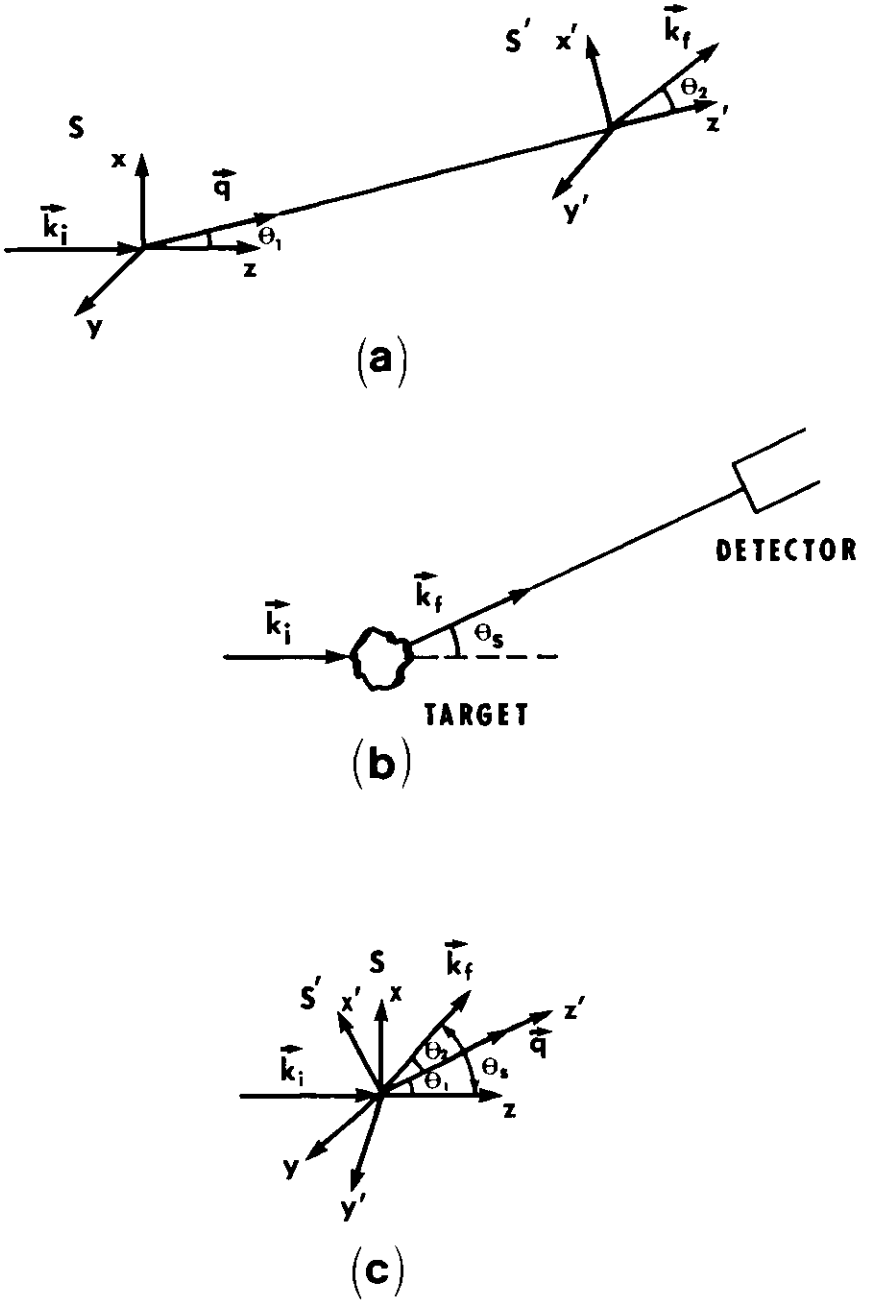

Figure 5. Target Geometry Used in Evaluating the Integral $G(k_i,\theta_S,\kappa_1)$.

APPENDIX IV

EVALUATION OF THE INTEGRAL - $\int d^3r\, e^{-i\vec{k}_1\cdot\vec{r}}\, \vec{\mu}_j\cdot\vec{\nabla}\left[\vec{\mu}_N\cdot\vec{\nabla}\left(\frac{1}{r}\right)\right]$

In this appendix, the integral appearing in Eq. (7.15) in the text is evaluated and shown to be

$$I \equiv -\int d^3r\, e^{-i\vec{k}_1\cdot\vec{r}}\, \vec{\mu}_j\cdot\vec{\nabla}\left[\vec{\mu}_N\cdot\vec{\nabla}\left(\frac{1}{r}\right)\right] = 4\pi(\vec{\mu}_N\cdot\hat{k}_1)(\vec{\mu}_j\cdot\hat{k}_1) \qquad \text{(IV.1)}$$

Letting

$$\phi(\vec{r}) \equiv \vec{\mu}_N\cdot\vec{\nabla}\left(\frac{1}{r}\right) \qquad \text{(IV.2)}$$

and

$$\vec{A} \equiv e^{-i\vec{k}_1\cdot\vec{r}}\, \vec{\mu}_j \qquad \text{(IV.3)}$$

and using the vector identity[19]

$$\vec{\nabla}\cdot(\phi\vec{A}) = \vec{A}\cdot\vec{\nabla}\phi + \phi\vec{\nabla}\cdot\vec{A} \qquad \text{(IV.4)}$$

The integral I is given by

$$I = \int d^3r\ \phi(\vec{r})\vec{\nabla}\cdot\vec{A} - \int d^3r\ \vec{\nabla}\cdot(\phi\vec{A}) \qquad \text{(IV.5)}$$

From the divergence theorem,[19] the second integral on the right-hand-side of Eq. (IV.5) can be written as

$$- \int d^3r \, \vec{\nabla}\cdot(\phi\vec{A}) = \int_S \phi\vec{A}\cdot d\vec{S} = \int_S e^{-i\vec{k}_1\cdot\vec{r}} \, \vec{\mu}_N\cdot\vec{\nabla}\left(\frac{1}{r}\right) \vec{\mu}_j\cdot d\vec{S} \qquad \text{(IV.6)}$$

where the limits of integration on the integral on the right-hand-side of Eq. (IV.6) are over the surface, S, bounding the (infinite) volume of integration of the integral on the left-hand-side of Eq. (IV.6). In the limit $S \to \infty$, the integral on the right-hand-side of Eq. (IV.6) vanishes as a consequence of the Riemann-Lebesque lemma.[43] Therefore, the integral I, from Eq. (IV.5), becomes

$$I = \int d^3r \, \phi(\vec{r})\vec{\nabla}\cdot\vec{A} = \int d^3r \, \vec{\mu}_N\cdot\vec{\nabla}\left(\frac{1}{r}\right)\vec{\nabla}\cdot\left[e^{-i\vec{k}_1\cdot\vec{r}} \, \vec{\mu}_j\right] \qquad \text{(IV.7)}$$

Now,

$$\vec{\nabla}\cdot\left[e^{-i\vec{k}_1\cdot\vec{r}} \, \vec{\mu}_j\right] = -i(\vec{\mu}_j\cdot\vec{k}_1) \, e^{-i\vec{k}_1\cdot\vec{r}} \qquad \text{(IV.8)}$$

Therefore,

$$I = -i(\vec{\mu}_j\cdot\vec{k}_1) \int d^3r \, e^{-i\vec{k}_1\cdot\vec{r}} \, \vec{\mu}_N\cdot\vec{\nabla}\left(\frac{1}{r}\right) \qquad \text{(IV.9)}$$

The integral I now has the same form as it did in Eq. (IV.1). Thus, now

letting

$$\phi(\vec{r}) \equiv \frac{1}{r} \tag{IV.10}$$

and

$$\vec{A} \equiv e^{-i\vec{k}\cdot\vec{r}}\,\vec{\mu}_N \tag{IV.11}$$

and following the same analysis as presented from Eq. (IV.4) to Eq. (IV.9) gives

$$I = (\vec{\mu}_N\cdot\vec{k}_1)(\vec{\mu}_j\cdot\vec{k}_1)\int d^3r\,\frac{e}{r}\,e^{-i\vec{k}_1\cdot\vec{r}} \tag{IV.12}$$

The integral in Eq. (IV.12) formally diverges but can be evaluated with the help of a convergence factor,

$$\int d^3r\,\frac{e}{r}\,e^{-i\vec{k}_1\cdot\vec{r}} \rightarrow \lim_{\alpha\to 0}\int d^3r\,\frac{e}{r}\,e^{-i\vec{k}_1\cdot\vec{r}}\,e^{-\alpha r}$$

$$= \lim_{\alpha\to 0}\left[\frac{4\pi}{k_1^2+\alpha^2}\right] = \frac{4\pi}{k_1^2} \tag{IV.13}$$

Therefore,

$$I = 4\pi(\vec{\mu}_N\cdot\hat{k}_1)(\vec{\mu}_j\cdot\hat{k}_1) \tag{IV.14}$$

where

$$\hat{k}_1 = \frac{\vec{k}_1}{k_1} \tag{IV.15}$$

which is Eq. (7.15) in the text.

APPENDIX V

THE ZERO-FIELD ISOTHERMAL SUSCEPTIBILITY FOR A FERROMAGNET AT $T > T_c$

In this appendix, the zero-field isothermal susceptibility for a ferromagnet in its paramagnetic state ($T > T_c$) is determined in terms of the spin-spin correlation function, as given by Eq. (7.69) in the text.

Consider a ferromagnet in its paramagnetic state ($T > T_c$) in the presence of a small external magnetic field H, and whose Hamiltonian is given by

$$H_{tot} = H - g_e \mu_B \vec{H} \cdot \vec{S}_T \tag{V.1}$$

where H is the Heisenberg Hamiltonian, Eq. (7.51), and $\vec{S}_T$ is the magnet's total spin, i.e.,

$$\vec{S}_T = \sum_j \vec{S}_j \tag{V.2}$$

Since the magnet is in its paramagnetic state, its mean magnetic moment, $\langle \vec{M} \rangle$, and the first derivative of its mean magnetic moment with respect to H will essentially be in the direction of $\vec{H}$. Therefore,

$$\frac{1}{H} \vec{H} \cdot \left(\frac{\partial \langle \vec{M} \rangle}{\partial H} \right)_T \cong \frac{1}{H} H \left(\frac{\partial \langle M \rangle}{\partial H} \right)_T = \left(\frac{\partial \langle M \rangle}{\partial H} \right)_T \equiv \chi_T(H) \tag{V.3}$$

where $\chi_T(H)$ is the isothermal susceptibility. The magnet's mean magnetic moment is given by

$$\langle \vec{M} \rangle = g_e \mu_B \langle \vec{S}_T \rangle = g_e \mu_B Z^{-1} \, tr\left(\vec{S}_T e^{-H_{tot}/k_B T} \right) \tag{V.4}$$

where Z, the partition function, is given by

$$Z = tr \, e^{-H_{tot}/k_B T} \tag{V.5}$$

and tr stands for trace. From Eq. (V.4),

$$\left(\frac{\partial \langle \vec{M} \rangle}{\partial H} \right)_T = g_e \mu_B \left\{ -Z^{-2} \, tr\left[\vec{S}_T \, e^{-H_{tot}/k_B T} \right] \left(\frac{\partial Z}{\partial H} \right)_T + Z^{-1} \, tr\left[\vec{S}_T \left(\frac{\partial e^{-H_{tot}/k_B T}}{\partial H} \right)_T \right] \right\} \tag{V.6}$$

From Eq. (V.1),

$$\left(\frac{\partial e^{-H_{tot}/k_B T}}{\partial H} \right)_T = \frac{g_e \mu_B}{k_B T} \, e^{-H_{tot}/k_B T} \, \frac{\vec{H} \cdot \vec{S}_T}{H} \tag{V.7}$$

thus,

$$\left(\frac{\partial \langle \vec{M} \rangle}{\partial H} \right)_T = \frac{g_e^2 \mu_B^2}{k_B T H} \left\{ \langle \vec{S}_T (\vec{H} \cdot \vec{S}_T) \rangle - \langle \vec{S}_T \rangle \langle \vec{H} \cdot \vec{S}_T \rangle \right\} \tag{V.8}$$

and

$$\chi_T(H) = \frac{1}{H}\vec{H}\cdot\left(\frac{\partial\langle\vec{M}\rangle}{\partial H}\right)_T = \frac{g_e^2\mu_B^2}{k_B T H^2}\left\{\langle(\vec{H}\cdot\vec{S}_T)^2\rangle - \langle\vec{H}\cdot\vec{S}_T\rangle^2\right\} \tag{V.9}$$

The zero-field isothermal susceptibility is defined as

$$\chi_T \equiv \lim_{H\to 0} \chi_T(H) \tag{V.10}$$

From Eq. (V.1), in the limit $H \to 0$,

$$H_{tot} = H \tag{V.11}$$

i.e., the total Hamiltonian is given by the Heisenberg Hamiltonian. Therefore, in this limit,

$$\langle(\vec{H}\cdot\vec{S}_T)^2\rangle = H^\alpha H^\beta \langle S_T^\alpha S_T^\beta\rangle = H^\alpha H^\beta \sum_{j,\ell} \langle S_j^\alpha S_\ell^\beta\rangle$$

$$= \frac{1}{3} H^2 \sum_{j,\ell} \langle\vec{S}_j\cdot\vec{S}_\ell\rangle \tag{V.12}$$

where the invariance to simultaneous rotation of the spins, Eq. (7.58), implied by the Heisenberg Hamiltonian has been used. Also, in the limit $H \to 0$

$$\langle\vec{H}\cdot\vec{S}_T\rangle^2 = H^\alpha H^\beta \langle S_T^\alpha\rangle\langle S_T^\beta\rangle = 0 \tag{V.13}$$

since $\langle S_T^\alpha \rangle = 0$ for a ferromagnet in its paramagnetic state in the absence of an external magnetic field. Thus, from Eq. (V.9).

$$T\chi_T = \frac{g_e^2 \mu_B^2}{3k_B} \sum_{j,\ell} \langle \vec{S}_j \cdot \vec{S}_\ell \rangle \tag{V.14}$$

For a translationally invariant spin system,

$$\sum_{j,\ell} \langle \vec{S}_j \cdot \vec{S}_\ell \rangle = N \sum_{\ell} \langle \vec{S}_0 \cdot \vec{S}_{\vec{R}_\ell} \rangle \tag{V.15}$$

Now,

$$N \sum_{\ell} \langle \vec{S}_0 \cdot \vec{S}_{\vec{R}_\ell} \rangle = N \sum_{\ell} \int d^3R \; \delta(\vec{R}_\ell - \vec{R}) \gamma(\vec{R})$$

$$= \frac{N}{(2\pi)^3} \sum_{\ell} \int d^3R \int d^3k_1 \; e^{i\vec{k}_1 \cdot (\vec{R}_\ell - \vec{R})} \gamma(\vec{R}) \tag{V.16}$$

where $\gamma(\vec{R}) = \langle \vec{S}_0 \cdot \vec{S}_{\vec{R}} \rangle$. Since there is only one atom per unit cell,

$$\sum_{\ell} e^{-i\vec{k}_1 \cdot \vec{R}_\ell} = (2\pi)^3 \rho \sum_{\tau} \delta(\vec{k}_1 - \vec{\tau}) \tag{V.17}$$

where τ is a reciprocal lattice vector. Therefore,

$$N \sum_{\ell} \langle S_0 \cdot S_{\vec{R}_\ell} \rangle = N\rho \sum_{\tau} \int d^3R \; e^{-i\vec{\tau}\cdot\vec{R}} \; \gamma(\vec{R}) \tag{V.18}$$

and

$$T\chi_T = \frac{Ng_e^2\mu_B^2}{3k_B} \rho \sum_{\tau} \int d^3R \; e^{-i\vec{\tau}\cdot\vec{R}} \; \gamma(\vec{R}) \tag{V.19}$$

which is Eq. (7.69) in the text.

APPENDIX VI

EVALUATION OF THE NEUTRON SPIN MATRIX ELEMENTS FOR DOUBLE SCATTERING

In this appendix, it is shown that, as given by Eq. (8.11) in the text,

$$M_N^{\alpha\beta\gamma\delta} \langle P_j^{\alpha}(\vec{k}_2) P_n^{\delta}(\vec{k}_4) \rangle \langle P_\ell^{\beta}(\vec{k}_1) P_m^{\gamma}(\vec{k}_3) \rangle$$

$$= \left(\frac{4\pi g e\hbar}{m_N c}\right)^4 f(\vec{k}_1,\vec{k}_2,\vec{k}_3,\vec{k}_4) \langle \vec{S}_j \cdot \vec{S}_n \rangle \langle \vec{S}_\ell \cdot \vec{S}_m \rangle \qquad \text{(VI.1)}$$

where, from Eq. (8.10),

$$M_N^{\alpha\beta\gamma\delta} = \frac{1}{2} \left(\frac{4\pi g e\hbar}{m_N c}\right)^4 \sum_{\sigma_i} \langle \sigma_i | s_N^{\alpha} s_N^{\beta} s_N^{\gamma} s_N^{\delta} | \sigma_i \rangle$$

$$\equiv \left(\frac{4\pi g e\hbar}{m_N c}\right)^4 \langle s_N^{\alpha} s_N^{\beta} s_N^{\gamma} s_N^{\delta} \rangle_N \qquad \text{(VI.2)}$$

from Eq. (8.5),

$$P_j^{\alpha}(\vec{k}_2) = \{\vec{S}_j - \hat{k}_2 (\vec{S}_j \cdot \hat{k}_2)\}^{\alpha} \qquad \text{(VI.3)}$$

and from Eq. (8.12),

$$f = \frac{1}{144}\left\{\left[1+(\hat{k}_2\cdot\hat{k}_4)^2\right]\left[1+(\hat{k}_1\cdot\hat{k}_3)^2\right] + (\hat{k}_2\cdot\hat{k}_4)(\hat{k}_1\cdot\hat{k}_3)(\hat{k}_2\times\hat{k}_4)\cdot(\hat{k}_1\times\hat{k}_3)\right\}_j$$

(VI.4)

Utilizing the invariance of the target spin system to simultaneous rotations of the spins, Eq. (7.58), it is easily shown that

$$\langle P_j^{\alpha}(\vec{k}_2)P_n^{\delta}(\vec{k}_4)\rangle = \frac{1}{3}\left[\delta_{\alpha,\beta} - \hat{k}_2^{\alpha}\hat{k}_2^{\delta} - \hat{k}_4^{\alpha}\hat{k}_4^{\delta} + \hat{k}_2^{\alpha}\hat{k}_4^{\delta}\,(\hat{k}_2\cdot\hat{k}_4)\right]\langle\vec{S}_j\cdot\vec{S}_n\rangle$$

(VI.5)

where $\hat{k}_2^{\alpha}$ denotes the α-component of the unit vector $\hat{k}_2$. Similarly,

$$\langle P_{\ell}^{\beta}(\vec{k}_1)P_m^{\gamma}(\vec{k}_3)\rangle = \frac{1}{3}\left[\delta_{\beta,\gamma} - \hat{k}_1^{\beta}\hat{k}_1^{\gamma} - \hat{k}_3^{\beta}\hat{k}_3^{\gamma} + \hat{k}_1^{\beta}\hat{k}_3^{\gamma}\,(\hat{k}_1\cdot\hat{k}_3)\right]\langle\vec{S}_{\ell}\cdot\vec{S}_m\rangle$$

(VI.6)

From Eq. (VI.2),

$$\langle s_N^{\alpha}s_N^{\beta}s_N^{\gamma}s_N^{\delta}\rangle_N = 2\ \mathrm{tr}(s_N^{\alpha}s_N^{\beta}s_N^{\gamma}s_N^{\delta})$$

$$= 2\ \mathrm{tr}(s_N^{\delta}s_N^{\alpha}s_N^{\beta}s_N^{\gamma}) = \langle s_N^{\delta}s_N^{\alpha}s_N^{\beta}s_N^{\gamma}\rangle_N \qquad \text{(VI.7)}$$

where the fact that the trace (tr) of a product of operators is invariant under a cyclic permutation of those operators has been used. From Eqs. (VI.2), (VI.5), (VI.6) and (VI.7), it is easily shown that

$$M_N^{\alpha\beta\gamma\delta} \langle P_j^{\alpha}(\vec{k}_2) P_n^{\delta}(\vec{k}_4) \rangle \langle P_\ell^{\beta}(\vec{k}_1) P_m^{\gamma}(\vec{k}_3) \rangle$$

$$= \left(\frac{4\pi g e\hbar}{m_N c}\right)^4 \frac{1}{9} \langle \vec{p}_N(\vec{k}_4)\cdot\vec{p}_N(\vec{k}_2)\vec{p}_N(\vec{k}_1)\cdot\vec{p}_N(\vec{k}_3) \rangle_N \langle \vec{S}_j\cdot\vec{S}_n \rangle \langle \vec{S}_\ell\cdot\vec{S}_m \rangle \quad \text{(VI.8)}$$

where $\vec{p}_N(\vec{k}_1)$ is the two-dimensional vector component of $\vec{s}_N$ perpendicular to $\hat{k}_1$, i.e.,

$$\vec{p}_N(\vec{k}_1) = \vec{s}_N - \hat{k}_1(\vec{s}_N\cdot\hat{k}_1) \quad \text{(VI.9)}$$

Comparing Eq. (VI.8) with Eq. (VI.1), one obtains

$$f = \frac{1}{9} \langle \vec{p}_N(\vec{k}_4)\cdot\vec{p}_N(\vec{k}_2)\vec{p}_N(\vec{k}_1)\cdot\vec{p}_N(\vec{k}_3) \rangle_N \quad \text{(VI.10)}$$

Notice, that if $\vec{k}_2 = \vec{k}_4$ and $\vec{k}_1 = \vec{k}_3$, then

$$\vec{p}_N(\vec{k}_4)\cdot\vec{p}_N(\vec{k}_2) = \vec{p}_N(\vec{k}_1)\cdot\vec{p}_N(\vec{k}_3) = p_N^2 = \frac{1}{2} \quad \text{(VI.11)}$$

a c-number, and

$$f = \frac{1}{36} \quad \text{(VI.12)}$$

in agreement with Eq. (8.18).

Obtaining f in the explicit form given by Eq. (VI.4) is now a straight-forward but tedious calculation. Substituting Eq. (VI.9) into

Eq. (VI.10), f can be written as a sum of averages in which appears, in every term, four neutron spin operators. Explicitly,

$$f = \frac{1}{9}\,[A^{(1)} + A^{(2)} + A^{(3)}] \tag{VI.13}$$

where

$$\begin{aligned} A^{(1)} = {} & \langle(\vec{s}_N\cdot\vec{s}_N)^2\rangle_N - \langle\vec{s}_N\cdot\vec{s}_N(\vec{s}_N\cdot\hat{k}_4)^2\rangle - \langle\vec{s}_N\cdot\vec{s}_N(\vec{s}_N\cdot\hat{k}_2)^2\rangle_N \\ & - \langle\vec{s}_N\cdot\vec{s}_N(\vec{s}_N\cdot\hat{k}_1)^2\rangle_N - \langle\vec{s}_N\cdot\vec{s}_N(\vec{s}_N\cdot\hat{k}_3)^2\rangle_N + \langle(\vec{s}_N\cdot\hat{k}_4)^2(\vec{s}_N\cdot\hat{k}_1)^2\rangle_N \\ & + \langle(\vec{s}_N\cdot\hat{k}_4)^2(\vec{s}_N\cdot\hat{k}_3)^2\rangle_N + \langle(\vec{s}_N\cdot\hat{k}_2)^2(\vec{s}_N\cdot\hat{k}_1)^2\rangle_N + \langle(\vec{s}_n\cdot\hat{k}_2)^2(\vec{s}_N\cdot\hat{k}_3)^2\rangle_N \end{aligned} \tag{VI.14}$$

$$\begin{aligned} A^{(2)} = {} & (\hat{k}_2\cdot\hat{k}_4)\langle\vec{s}_N\cdot\vec{s}_N\ \vec{s}_N\cdot\hat{k}_4\ \vec{s}_N\cdot\hat{k}_2\rangle_N + (\hat{k}_1\cdot\hat{k}_3)\langle\vec{s}_N\cdot\vec{s}_N\ \vec{s}_N\cdot\hat{k}_1\ \vec{s}_N\cdot\hat{k}_3\rangle_N \\ & - (\hat{k}_2\cdot\hat{k}_4)\langle(\vec{s}_N\cdot\hat{k}_1)^2\vec{s}_N\cdot\hat{k}_4\ \vec{s}_N\cdot\hat{k}_2\rangle_N - (\hat{k}_2\cdot\hat{k}_4)\langle(\vec{s}_N\cdot\hat{k}_1)^2\vec{s}_N\cdot\hat{k}_4\ \vec{s}_N\cdot\hat{k}_2\rangle_N \\ & - (\hat{k}_1\cdot\hat{k}_3)\langle(\vec{s}_N\cdot\hat{k}_4)^2\vec{s}_N\cdot\hat{k}_1\ \vec{s}_N\cdot\hat{k}_3\rangle_N - (\hat{k}_1\cdot\hat{k}_3)\langle(\vec{s}_N\cdot\hat{k}_2)^2\vec{s}_N\cdot\hat{k}_1\ \vec{s}_N\cdot\hat{k}_3\rangle_N \end{aligned} \tag{VI.15}$$

$$A^{(3)} = (\hat{k}_2\cdot\hat{k}_4)(\hat{k}_1\cdot\hat{k}_3)\langle\vec{s}_N\cdot\hat{k}_4\ \vec{s}_N\cdot\hat{k}_2\ \vec{s}_N\cdot\hat{k}_1\ \vec{s}_N\cdot\hat{k}_3\rangle_N \tag{VI.16}$$

Each of these averages can be evaluated by noting that

$$\langle s_N^{\alpha} s_N^{\beta} s_N^{\gamma} s_N^{\delta} \rangle_N = \frac{1}{16} (\delta_{\alpha,\delta}\delta_{\beta,\gamma} - \delta_{\alpha,\gamma}\delta_{\beta,\delta} + \delta_{\alpha,\beta}\delta_{\gamma,\delta}) \tag{VI.17}$$

For example, $A^{(3)}$ can be written as

$$A^{(3)} = (\hat{k}_2 \cdot \hat{k}_4)(\hat{k}_1 \cdot \hat{k}_3) \langle s_N^{\alpha} s_N^{\beta} s_N^{\gamma} s_N^{\delta} \rangle_N \hat{k}_4^{\alpha} \hat{k}_2^{\beta} \hat{k}_1^{\gamma} \hat{k}_3^{\delta} \tag{VI.18}$$

Thus, from Eq. (VI.17),

$$A^{(3)} = \frac{1}{16} (\hat{k}_2 \cdot \hat{k}_4)(\hat{k}_1 \cdot \hat{k}_3)(\varepsilon_{\alpha\beta\nu}\varepsilon_{\delta\gamma\nu} + \delta_{\alpha,\beta}\delta_{\gamma,\delta}) \hat{k}_4^{\alpha} \hat{k}_2^{\beta} \hat{k}_1^{\gamma} \hat{k}_3^{\delta} \tag{VI.19}$$

where the identity[44]

$$\varepsilon_{\alpha\beta\nu}\varepsilon_{\delta\gamma\nu} = \delta_{\alpha,\delta}\delta_{\beta,\gamma} - \delta_{\alpha,\gamma}\delta_{\beta,\delta} \tag{VI.20}$$

has been used, and where $\varepsilon_{\alpha\beta\nu}$ is the permutation symbol.[45] Thus,

$$A^{(3)} = \frac{1}{16} (\hat{k}_2 \cdot \hat{k}_4)(\hat{k}_1 \cdot \hat{k}_3) \left[(\hat{k}_2 \times \hat{k}_4) \cdot (\hat{k}_1 \times \hat{k}_3) + (\hat{k}_2 \cdot \hat{k}_4)(\hat{k}_1 \cdot \hat{k}_3) \right] \tag{VI.21}$$

where the identity[46]

$$\varepsilon_{\alpha\beta\nu}\varepsilon_{\delta\gamma\nu} \hat{k}_4^{\alpha} \hat{k}_2^{\beta} \hat{k}_1^{\gamma} \hat{k}_3^{\delta} = (\hat{k}_2 \times \hat{k}_4) \cdot (\hat{k}_1 \times \hat{k}_3) \tag{VI.22}$$

has been used.

Similarly, it can be shown that

$$A^{(1)} = \frac{1}{16} \tag{VI.23}$$

and

$$A^{(2)} = \frac{1}{16}\left[(\hat{k}_2\cdot\hat{k}_4)^2 + (\hat{k}_1\cdot\hat{k}_3)^2\right] \tag{VI.24}$$

Therefore,

$$f = \frac{1}{144}\Big[1+(\hat{k}_2\cdot\hat{k}_4)^2+(\hat{k}_1\cdot\hat{k}_3)^2+(\hat{k}_2\cdot\hat{k}_4)(\hat{k}_1\cdot\hat{k}_3)(\hat{k}_2\times\hat{k}_4)\cdot(\hat{k}_1\times\hat{k}_3) + (\hat{k}_2\cdot\hat{k}_4)^2(\hat{k}_1\cdot\hat{k}_3)^2\Big] \tag{VI.25}$$

or

$$f = \frac{1}{144}\left\{\left[1+(\hat{k}_2\cdot\hat{k}_4)^2\right]\left[1+(\hat{k}_1\cdot\hat{k}_3)^2\right] + (\hat{k}_2\cdot\hat{k}_4)(\hat{k}_1\cdot\hat{k}_3)(\hat{k}_2\times\hat{k}_4)\cdot(\hat{k}_1\times\hat{k}_3)\right\} \tag{VI.26}$$

which is identical to Eq. (VI.4).

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9. In solving the scattering problem via this method, $\psi(\vec{r},t)$ for all times t must, of course, satisfy $(H_p + \phi)\psi(\vec{r},t) = i\hbar \frac{\partial\psi(\vec{r},t)}{\partial t}$ where ϕ is the interaction between the probe and the target. Formally, Eq. (2.11) is such a solution if $\psi_k(\vec{r})$ is an eigenfunction of $H_p + \phi$. This approach is considered in detail for a fixed scattering center by E. Merzbacher, Quantum Mechanics, 2nd Ed. (Wiley, New York, 1970), Chap. 11.

10. The dependence of S on two wave vectors $\vec{k}_2$, $\vec{k}_4$ rather than the single wave vector dependence of $S(\vec{k},\omega)$ of the first Born approximation, Eq. (4.26), is a consequence of recognizing the finite target size for multiple scattering. For a truly translational invariant (infinite) target, $\vec{k}_2 = -\vec{k}_4$, and S would depend only on a single wave vector (see Chapter VI).

11. Possible gravitational effects that can cause a mean density gradient in the liquid are neglected, i.e., it is assumed that Eq. (5.17) is valid.

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16. Retaining the self-correlation term in $\Gamma(r)$ results in three additional terms for the double scattering critical nuclear DCS in addition to the one given in Eq. (6.37). These three additional terms are all independent of k; thus they represent isotropic scattering terms and are of little interest.

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21. For example, at iron's Curie temperature, $T_c = 1043^o$ K, the amount of thermal energy available per atom is $\sim k_B T_c = 0.09$ eV, which is much less than the energy needed ($\sim$ 1 eV) to cause an electronic excitation.

22. Thermal-neutrons have energies on the order of $10^{-3} - 10^{-1}$ eV, which is much less than the energy needed ($\sim$ 1 eV) to cause an electronic excitation.

23. The Heisenberg spin quantum number, S_j, may have to be adjusted to account for electronic orbital angular momentum, crystalline field effects, etc. For example, iron has four (d orbital) unpaired electrons; yet the effective Heisenberg spin for iron is not two but approximately 1.1.

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28. For references of critical light experiments, see article by M. E. Fisher, Reference 7.

29. D. W. Oxtoby and W. M. Gelbart, J. Chem. Phys. 60, 3359 (1974); Phys. Rev. A 10, 738 (1974).

30. See, for example, Handbook of Mathematical Functions, edited by M. Abramowitz and I. A. Stegun (U.S. Dept. Comm., Washington, D.C., 1966), Chap. 5.

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34. In the treatment given by Glauber, Reference 8, $\vec{r}_j$ is replaced by the component of $\vec{r}_j$ perpendicular to $\vec{k}_i$; however, whether one uses this component or $\vec{r}_j$ itself makes no difference in any of the results.

35. The two dimensional integral over $\vec{\lambda}$ is evaluated by letting $\vec{k}$ lie along the x-axis in $\vec{\lambda}$-space, using the small-angle approximation for k, i.e., $k = k_i\theta_S$ and using the integrals 2.553(3), p. 148 and 2.266, p. 84 of Reference 31.

36. To establish the equivalency of these two expressions for $\langle \ell \rangle$, one must bear in mind that the $\ell(\vec{b})$ in Eq. (10.50) is defined as the length of the target parallel to $\vec{k}_i$ and going through the point $\vec{b}$, while $\ell(\vec{r})$ in Eq. (6.36) is the length from point r in the direction of $\vec{k}_i$ to the boundary of the target.

37. Reference 31, integrals 8.411(1), p. 952, and 6.532(4), p. 678.

REFERENCES (concluded)

38. For small z, $K_0(z)$ goes like $\ell n(2/z)$. Substituting this in Eq. (10.70), the integral is made convergent by means of a convergence factor, exp $[-\alpha b'']$. Thus, from integral 6.621(1), p. 711, of Reference 31, Eq. (10.71) is obtained in the limit $\alpha \to 0$.

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41. J. B. Marion, Classical Dynamics of Particles and Systems, 2nd Ed. (Academic, New York, 1970), p. 384.

42. Reference 31, integrals 2.553(3), p. 148 and 2.266, p. 84.

43. J. T. Cushing, Applied Analytical Mathematics for Physical Scientists, (Wiley, New York, 1975), p. 158.

44. Reference 41, p. 25, Eq. 5.

45. Reference 41, p. 23.

46. Reference 41, p. 25, Eq. 4.

VITA

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